



**REPUBLIC OF TURKEY
ADANA ALPARSLAN TÜRKEŞ SCIENCE AND TECHNOLOGY
UNIVERSITY**

**INSTITUTE OF GRADUATE STUDIES
DEPARTMENT OF MATERIALS ENGINEERING**

**INVESTIGATION OF ADVANCED RADAR ABSORBER COMPOSITE
MATERIALS HAVING HIGH STEALTH TECHNOLOGY TO USE IN
VARIOUS MILITARY VEHICLES**

**KUTLUHAN UTKU TÜMEN MASTER
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**SUPERVISOR
ASSOC. PROF. DR. MUSTAFA AKYOL**

ADANA 2021

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MILITARY VEHICLES**

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02/07/2021





I hereby declare that all information in this thesis has been obtained and presented in accordance with academic rules and ethical conduct. I also declare that, as required by these rules and conduct, I have fully cited and referenced all information that is not original to this work.

Kutluhan Utku TMEN

ABSTRACT

INVESTIGATION OF ADVANCED RADAR ABSORBER COMPOSITE MATERIALS HAVING HIGH STEALTH TECHNOLOGY TO USE IN VARIOUS MILITARY VEHICLES

Kutluhan Utku TÜMEN

Department of Materials Engineering

Supervisor: Assoc. Prof. Dr. Mustafa AKYOL July

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The aim of the stealth technology, which has attracted great attention in the field of defense technologies in recent years, is to reduce the radar cross-sectional area (RCS) of the target object. Reducing the RCS can prevent the potential target object from being seen in enemy radar systems. Radar absorber materials are material groups that actively exhibit the stealth technology by losing the visibility of the target object by reducing and absorbing the energy of electromagnetic waves entering the material with the magnetic and dielectric loss mechanism. In this study, $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ ($0 \leq x \leq 1$ with increments 0.25) and $\text{Ni}_y\text{:ZnO}$ ($y=0$ and 0.1) nanoparticles were synthesized using the sol-gel method. X-Ray Diffraction (XRD), Scanning Electron Microscope (SEM), Energy Distributed X-Ray Spectroscopy (EDS) and Vibrating Sample Magnetometer (VSM) analyzes were performed for determining the crystal structures, particle size and morphology, elemental analysis and magnetic characteristics of the produced nanoparticles, respectively. Various radar absorbing composite structures were obtained by using the epoxy casting method with the addition of the produced nanoparticles into the epoxy resin. Microwave absorption properties of nanoparticle reinforced epoxy matrix composite structures were characterized in the 8-12 GHz (X band) frequency range using a Vector Network Analyzer. In addition to the mechanical properties of composites were evaluated by applying hardness and impact test. It has been observed that the microwave absorption and mechanical properties of the nanoparticle reinforced epoxy matrix composite samples are satisfactory and can be used in potential military land, air and sea applications.

Keywords: Stealth technology, radar cross section, sol-gel, microwave absorption.

ÖZET

YÜKSEK GİZLİLİK TEKNOLOJİSİNE SAHİP GELİŞMİŞ RADAR SOĞURUCU KOMPOZİT MALZEMELERİN ÇEŞİTLİ ASKERİ ARAÇLARDA KULLANILMASI

İÇİN ARAŞTIRILMASI

Kutluhan Utku TÜMEN

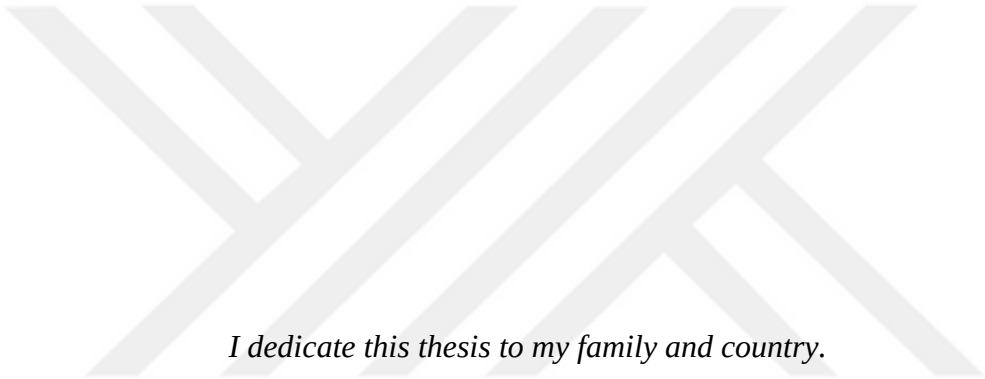
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Son yıllarda savunma teknolojileri alanında yoğun ilgi gören gizlilik teknolojisinin amacı, hedefteki nesnenin radar kesit alanını (RCS) azaltmaktır. RCS'yi azaltmak, potansiyel hedef olan nesnenin düşman radar sistemlerinde görünürlüğünü engelleyebilmektedir. Radar soğurucu malzemeler (RAM) manyetik ve dielektrik kayıp mekanizması ile malzeme içerisine giren elektromanyetik dalgaların enerjisini azaltarak ve soğurarak hedefteki nesnenin görünürlüğünü kaybedip gizlilik teknolojisini aktif olarak sergileyen malzeme gruplarıdır. Bu çalışmada sol-jel yöntemi kullanılarak $Ni_xCo_{1-x}Fe_2O_4$ ($0 \leq x \leq 1$, 0.25 artışla) ve $Ni_y:ZnO$ ($y=0$ ve 0.1) nanoparçacıkları sentezlenmiştir. Üretilen nanoparçacıkların kristal yapıları, partikül boyut ve morfolojisi, elementel analizi ve manyetik karakteristiklerinin belirlenmesi için sırasıyla X-Işını Kırınımı (XRD), Taramalı Elektron Mikroskobu (SEM), Enerji Dağıtım X-Işını Spektroskopisi (EDS) ve Titreşimli Örnek Manyetometresi (VSM) ölçümleri gerçekleştirilmiştir. Üretilen nanoparçacıkların epoksi reçine içerisine katkılanması ile epoksi döküm yöntemi kullanılarak çeşitli radar soğurucu kompozit yapıları elde edilmiştir. Nanoparçacık takviyeli epoksi matrisli kompozit yapılarının mikrodalga soğurma özellikleri, Vektörel Ağ Analizörü kullanılarak 8-12 GHz (X bandı) frekans aralığında karakterize edilmiştir. Ayrıca, sertlik ve darbe testi uygulanarak kompozitlerin mekanik özellikleri incelenmiştir. Nanoparçacık takviyeli epoksi matrisli kompozit numunelerin mikrodalga absorblama ve mekanik özelliklerinin tatmin edici düzeyde olduğu görülerek potansiyel askeri kara, hava ve deniz uygulamalarında kullanılabileceği görülmüştür.

Anahtar Kelimeler: Gizlilik teknolojisi, radar kesit alanı, sol-jel, mikrodalga soğurma.



I dedicate this thesis to my family and country.

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TABLE OF CONTENTS

TABLE OF CONTENTS	viii
LIST OF FIGURES	xi
LIST OF TABLES	xiv
NOMENCLATURE	xv
1. INTRODUCTION	1
1.1. Electromagnetic wave basics and absorbing theory	2
1.1.1. Dielectric mechanism	5
1.1.2. Magnetic mechanism	5
1.1.3. Loss tangent ($\tan\delta$)	6
1.1.4. Radar absorbing theory	6
1.2. Radar absorbing structures	8
1.2.1. Shape of military vehicles	9
1.2.2. Radar absorbing materials	11
1.2.2.1. Dielectric absorbers	11
1.2.2.2. Magnetic absorbers	11
1.2.2.3. Oxide absorbers	12
1.2.2.4. Polymeric composites	12
1.3. Magnetism	12
1.3.1. Origins of magnetism	13
1.3.2. Classification of magnetic materials	15
1.3.2.1. Diamagnetism	15
1.3.2.2. Paramagnetism	17

1.3.2.3. Ferromagnetism	18
1.3.2.4. Antiferromagnetism	20
1.3.2.5. Ferrimagnetism	21
1.3.3. Curie temperature and Curie-Weiss law	22
1.3.4. Magnetic hysteresis loop	24
1.4. Thesis objectives	24
2. LITERATURE REVIEW	26
3. MATERIALS AND METHODS	31
3.1. Sol-gel process	31
3.2. Preparation of nanomaterials	32
3.2.1. Synthesis of $\text{Ni}_y\text{:ZnO}$ nanoparticles	32
3.2.2. Synthesis of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ nanoparticles	33
3.2.3. Heat treatments	34
3.2.4. Production of radar absorbing composites (RAC)	36
3.2.5. Composite sample design for mechanical tests	38
3.3. Characterization techniques	40
3.3.1. Structural analysis	40
3.3.1.1. X-ray diffraction (XRD) method	40
3.3.1.2. Scanning electron microscopy (SEM) and energy-dispersive x-ray spectrum (EDS) methods	42
3.3.2. Magnetic analysis	43
3.3.3. Electromagnetic analysis	44
3.3.4. Mechanical analysis	45
3.3.4.1. Charpy impact test	45
3.3.4.2. Hardness test	46
4. RESULTS AND DISCUSSIONS	47
4.1. Structural properties of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ ($0.0 \leq x \leq 1.0$) and $\text{Ni}_y\text{:ZnO}$ ($y=0$ and 0.1) nanoparticles	47
4.1.1. X-ray diffraction (XRD) analysis of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ ($0.0 \leq x \leq 1.0$) and $\text{Ni}_y\text{:ZnO}$	

($y=0$ and 0.1) nanoparticles	47
4.1.2. Scanning electron microscope (SEM) and energy-dispersive x-ray spectrum (EDS) analysis of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ ($0.0 \leq x \leq 1.0$) and $\text{Ni}_y\text{:ZnO}$ ($y=0$ and 0.1) nanoparticles	52
4.2. Magnetic properties of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ ($0.0 \leq x \leq 1.0$) and $\text{Ni}_y\text{:ZnO}$ ($y=0$ and 0.1) nanoparticles	58
4.2.1. Temperature dependence of magnetic properties of $\text{Ni}_y\text{:ZnO}$ nanoparticles	58
4.2.2. Magnetic hysteresis of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ and $\text{Ni}_y\text{:ZnO}$ nanoparticles	60
4.3. Microwave absorption properties of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ and $\text{Ni}_y\text{:ZnO}$ nanoparticles reinforced epoxy matrix composites	67
4.4. Mechanical properties of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ and $\text{Ni}_y\text{:ZnO}$ nanoparticles reinforced epoxy matrix radar absorbing composite structures.....	76
4.4.1. Charpy impact test	76
4.4.2. Hardness test	76
5. CONCLUSIONS	78
6. RECOMMENDATIONS	81
REFERENCES	82
CURRICULUM VITAE	86

LIST OF FIGURES

Figure 1.1. The working principle of radar system.	1
Figure 1.2. a) Creation of electromagnetic waves (Verhoeven, 2017) b) Electromagnetic spectrum (Callister & Rethwisch, 2010).	3
Figure 1.3. Microwave spectrum with increasing frequencies (Hu, 2010).	4
Figure 1.4. Design of a) a passenger plane and b) an aircraft.	9
Figure 1.5. First generation aircrafts with stealth technology. a) F-117 Nighthawk b) B-2 Spirit Bomber and c) Blackbird (Blackbird; Nighthawk; Spirit).	9
Figure 1.6. Latest generation aircrafts with stealth technology. a) Lockheed Martin F-22 Raptor b) Sukhoi Su-57 c) Lockheed Martin F-35 Lightning II (LockheedMartinF-22Raptor; LockheedMartinF-35Lightning-II; SukhoiSu-57).	10
Figure 1.7. Warships with stealth technology. a) Sea Shadow (IX-529) b) HSwMS Helsingborg (K32) c) Kolkata-class destroyer (HSwMSHelsingborg(K32); Kolkata-classdestroyer; SeaShadow(IX-529)).	10
Figure 1.8. Ground war vehicles with stealth technology. a) PL-01 b) Boxer c) Challenger 2 (Boxer; Challenger2; PL-01).	11
Figure 1.9. Magnetic moment generation by current loop of an electron rotating around the nucleus with velocity V at radius R (Akyol, 2011).	13
Figure 1.10. Arrangement of magnetic moments of the material exhibiting diamagnetic behavior: a) external magnetic field is not applied, b) external magnetic field is applied and c) external magnetic field is removed.	16
Figure 1.11. $M-H$ and $\chi - T$ graphs of diamagnetic materials (Akyol, 2011).	16
Figure 1.12. Arrangement of magnetic moments of the material exhibiting paramagnetic behavior: a) external magnetic field is not applied, b) external magnetic field is applied and c) external magnetic field is removed.	17
Figure 1.13. $M-H$ and $\chi - T$ graphs of paramagnetic materials (Akyol, 2011).	18
Figure 1.14. Arrangement of magnetic moments of the material exhibiting ferromagnetic behavior: a) external magnetic field is not applied and b) external magnetic field is applied.	19
Figure 1.15. a) $M-H$ and $\chi - T$ graphs of ferromagnetic materials. Hysteresis curves of b) hard and c) soft ferromagnetic materials (Akyol, 2011).	19
Figure 1.16. Arrangement of magnetic moments of the material exhibiting antiferromagnetic behavior: a) external magnetic field is not applied and b) external magnetic field is applied.	20
Figure 1.17. $M-H$ graph of ferromagnetic materials (Akyol, 2011).	21
Figure 1.18. Arrangement of magnetic moments of the material exhibiting ferrimagnetic behavior: a) external magnetic field is not applied and b) external magnetic field is applied.	21
Figure 1.19. $M-H$ and $\chi - T$ graph of ferrimagnetic materials (Akyol, 2011).	22
Figure 1.20. Magnetic susceptibility curves of ferromagnetic, antiferromagnetic and	

paramagnetic materials (Akyol, 2011).	23
Figure 1.21. Hysteresis loop of a multi-domain magnetic material (Akyol, 2011).	24
Figure 2.1. Reflection loss versus frequency graph of nanocomposites (Kumar Chakradhary & Akhtar, 2021).	30
Figure 3.1. Operation scheme of the general sol-gel method.	32
Figure 3.2. Sol-gel process of $\text{Ni}_y\text{:ZnO}$ nanoparticles.	33
Figure 3.3. Sol-gel process of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ nanoparticles.	34
Figure 3.4. Diagram of the three stage heat treatment process: a) burning, b) calcination and c) sintering.	35
Figure 3.5. General production scheme of nanoparticle doped epoxy matrix composite.	36
Figure 3.6. Shape and size scheme of composite structures.	37
Figure 3.7. Step-by-steps from nanomaterial production to composite material production. .	38
Figure 3.8. Impact test specimen dimensions in accordance with ASTM D256 standards. ...	38
Figure 3.9. Hardness test specimen dimensions in accordance with ASTM D785 standards. 39	
Figure 3.10. Mold samples prepared from wood and epoxy casting for mechanical tests.	39
Figure 3.11. X-ray diffraction and Bragg's law on crystal planes ("Bragg Law," 2020).	41
Figure 3.12. X-ray diffraction system of PANalytical EMPYREAN.	41
Figure 3.13. Scanning electron microscope working principle scheme (SEM, 2020).	42
Figure 3.14. FEI Quanta FEG-650 type scanning electron microscope with EDS integrated. 43	
Figure 3.15. Quantum Design Dyna-Cool model Physical Property Measurement Sytem (PPMS).	44
Figure 3.16. PNA-L type KEYSIGHT network analyzer system.	45
Figure 3.17. MITECH XJJ-50S digital charpy impact tester.	46
Figure 3.18. LOYKA Digital Shore D Durometer hardness test analyzer.	46
Figure 4.1. X-ray diffraction patterns of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ nanoparticles for a) $x=0.00$ b) $x=0.25$ c) $x=0.50$ d) $x=0.75$ and e) $x=1.00$	48
Figure 4.2. Lattice parameter (a) as function of Ni-content in $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ samples.	49
Figure 4.3. X-ray diffraction patterns of $\text{Ni}_y\text{:ZnO}$ nanoparticles for a) $y=0.00$ and b) $y=0.10$	51
Figure 4.4. CNF0 sample; a-b) SEM images, c) particle size distribution plot and d) EDS spectrum (inset of d, atomic and weight percentage of elements).	53
Figure 4.5. CNF1 sample; a-b) SEM images, c) particle size distribution plot and d) EDS spectrum (inset of d, atomic and weight percentage of elements).	53
Figure 4.6. CNF2 sample; a-b) SEM images, c) particle size distribution plot and d) EDS spectrum (inset of d, atomic and weight percentage of elements).	54
Figure 4.7. CNF3 sample; a-b) SEM images, c) particle size distribution plot and d) EDS spectrum (inset of d, atomic and weight percentage of elements).	54
Figure 4.8. CNF4 sample; a-b) SEM images, c) particle size distribution plot and d) EDS spectrum (inset of d, atomic and weight percentage of elements).	55
Figure 4.9. Average particle size as a function of Ni amount in $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ structure.	

.....	55	Figure 4.10. ZN0 sample; a-b) SEM images, c) particle size distribution plot and d) EDS spectrum (inset of d, atomic and weight percentage of elements).
.....	57	Figure 4.11. ZN1 sample; a-b) SEM images, c) particle size distribution plot and d) EDS spectrum (inset of d, atomic and weight percentage of elements).
.....	57	Figure 4.12. Temperature dependent magnetic moment curves of Ni_yZnO for $y=0$.
	59	
		Figure 4.13. Temperature dependent magnetic moment curves of Ni_yZnO for $y=0.1$.
	59	
		Figure 4.14. Magnetic hysteresis curve of CoFe_2O_4 at 300 K.
	60	
		Figure 4.15. Magnetic hysteresis curve of $\text{Ni}_{0.25}\text{Co}_{0.75}\text{Fe}_2\text{O}_4$ at 300 K.
	61	
		Figure 4.16. Magnetic hysteresis curve of $\text{Ni}_{0.5}\text{Co}_{0.5}\text{Fe}_2\text{O}_4$ at 300 K.
	61	
		Figure 4.17. Magnetic hysteresis curve of $\text{Ni}_{0.75}\text{Co}_{0.25}\text{Fe}_2\text{O}_4$ at 300 K.
	62	
		Figure 4.18. Magnetic hysteresis curve of NiFe_2O_4 at 300 K.
	62	
		Figure 4.19. a) Coercive field (H_c), b) saturation magnetization (M_s), c) remanence magnetization (M_r) and d) the ratio of squareness (M_r/M_s) of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ samples at 300K.
.....		
	64	
		Figure 4.20. Magnetic hysteresis curve of ZnO at 5 K.
	65	
		Figure 4.21. Magnetic hysteresis curve of ZnO at 300 K.
	65	
		Figure 4.22. Magnetic hysteresis curve of $\text{Ni}_{0.1}\text{ZnO}$ at 5 K.
	66	
		Figure 4.23. Magnetic hysteresis curve of $\text{Ni}_{0.1}\text{ZnO}$ at 300 K.
	66	
		Figure 4.24. Reflection loss plot of the pure epoxy resin without nanoparticles.
	68	
		Figure 4.25. Reflection loss plot of the 15% weight ZnO+15% weight CoFe_2O_4 nanoparticles reinforced epoxy RAC1 composite.
	69	
		Figure 4.26. Reflection loss plot of the 15% weight $\text{Ni}_{0.1}\text{ZnO}$ +15% weight CoFe_2O_4 nanoparticles reinforced epoxy RAC2 composite.
	69	
		Figure 4.27. Reflection loss plot of the 15% weight $\text{Ni}_{0.1}\text{ZnO}$ +15% weight $\text{Ni}_{0.25}\text{Co}_{0.75}\text{Fe}_2\text{O}_4$ nanoparticles reinforced epoxy RAC3 composite.
	70	
		Figure 4.28. Reflection loss plot of the 15% weight $\text{Ni}_{0.1}\text{ZnO}$ +15% weight $\text{Ni}_{0.5}\text{Co}_{0.5}\text{Fe}_2\text{O}_4$ nanoparticles reinforced epoxy RAC4 composite.
	71	
		Figure 4.29. Reflection loss plot of the 15% weight $\text{Ni}_{0.1}\text{ZnO}$ +15% weight $\text{Ni}_{0.75}\text{Co}_{0.25}\text{Fe}_2\text{O}_4$ nanoparticles reinforced epoxy RAC5 composite.
	71	
		Figure 4.30. Reflection loss plot of the 15% weight $\text{Ni}_{0.1}\text{ZnO}$ +15% weight NiFe_2O_4 nanoparticles reinforced epoxy RAC6 composite.
	72	
		Figure 4.31. Reflection loss plot of the 15% weight ZnO+15% weight $\text{Ni}_{0.25}\text{Co}_{0.75}\text{Fe}_2\text{O}_4$ nanoparticles reinforced epoxy RAC7 composite.
	73	
		Figure 4.32. Reflection loss plot of the 15% weight ZnO+15% weight $\text{Ni}_{0.5}\text{Co}_{0.5}\text{Fe}_2\text{O}_4$ nanoparticles reinforced epoxy RAC8 composite.
	73	
		Figure 4.33. Reflection loss plot of the 15% weight ZnO+15% weight $\text{Ni}_{0.75}\text{Co}_{0.25}\text{Fe}_2\text{O}_4$ nanoparticles reinforced epoxy RAC9 composite.
	74	
		Figure 4.34. Reflection loss plot of the 15% weight ZnO+15% weight NiFe_2O_4 nanoparticles reinforced epoxy RAC10 composite.
	75	
		Figure 4.35. Locations of hardness values taken from different places on the sample surface.

LIST OF TABLES

Table 1.1. Usage areas of radar frequencies in different bands (Bhatta & Geethapriya, 2016).	5
Table 3.1. The abbreviations used in nanomaterials and composite structures.	37
Table 4.1. Structural properties of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ samples.	50
Table 4.2. Structural properties of $\text{Ni}_y\text{:ZnO}$ samples.	52
Table 4.3. The desired and EDS results of atomic percentage of each element in $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ samples.	56
Table 4.4. The desired and EDS results of atomic percentage of each element in $\text{Ni}_y\text{:ZnO}$ samples.	58
Table 4.5. Coercive field (H_c), saturation magnetizations (M_s), remenant magnetization (M_r) and squareness ratio (M_r/M_s) values of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ samples.	63
Table 4.6. Coercive field (H_c), saturation magnetization (M_s), remenant magnetization (M_r) and squareness ratio (M_r/M_s) values of $\text{Ni}_y\text{:ZnO}$	67
Table 4.7. The absorption values of composite structures.	76
Table 4.8. Hardness values taken from five different point on the sample.	77

NOMENCLATURE

$\text{\AA}$	: Angstrom
$a^{\rightarrow}$	: Acceleration
$B^{\rightarrow\rightarrow}$	: Magnetic field
C	: Curie constant
$^{\circ}\text{C}$	: Celsius degree
D	: Crystallite size
$E^{\rightarrow\rightarrow}$	: Electric field
e^{-}	: Electron charge
EDS	: Energy-Dispersive X-Ray Spectroscopy
eV	: Electron Volts
FC	: Field Cooled
GHz	: Gigahertz
g	: Gram
hkl	: Miller indices
$H^{\rightarrow\rightarrow}$	: External magnetic field
H_c	: Coercive field
I	: Current
J	: Current density
$J^{\rightarrow}$	: Total angular momentum
j	: Total angular momentum quantum number
K	: Kelvin
k_B	: Boltzmann Constant
$L^{\rightarrow\rightarrow}$	: Orbital angular momentum
l	: Orbital angular momentum quantum number
MHz	: Megahertz
m	: Mass
$M^{\rightarrow\rightarrow\rightarrow}$	: Magnetization vector

M_s	: Saturation Magnetization
M_r	: Remanence Magnetization
Oe	: Oersted
PPMS	: Physical Properties Measurement System
R	: Radius
RAC	: Radar Absorbing Composites
RAM	: Radar Absorbing Materials
RCS	: Radar Cross Section
RL	: Reflection Loss
RF	: Radio Frequency
$S^{\rightarrow}$	: Spin angular momentum quantum number
SEM	: Scanning Electron Microscopy
T	: Temperature
T_B	: Blocking temperature
T_C	: Curie temperature
T_N	: Neel temperature
VSM	: Vibrating Sample Magnetometer
ZFC	: Zero Field Cooled
XRD	: X-Ray Diffraction
ϵ	: Permittivity
μ	: Permeability
Q	: Temperature constant
τ	: Relaxation time
V	: Speed
χ	: Magnetic susceptibility
λ	: Wavelength
θ	: Bragg angle
μ	: Magnetic moment
μ_B	: Bohr magnetron
μ_0	: Free space magnetic permeability
γ	: Gyromagnetic ratio
π	: Pi number

1. INTRODUCTION

Radar, which occupies an important place in the defense industry, has been gained more importance due to occurring extraordinary progress with the development of weapon technology since the second world war. As a result of the development in radar technology, a revolution has occurred for weapon systems in air, land and sea. The term RADAR was defined by the United States Navy in 1940 as an acronym for RAdio, DeTection and RAnging (Knott, Shaeffer, & Tuley, 2004).

Radar is a technological system that helps to find the height, speed, altitude, range and direction of fixed and moving objects with the help of electromagnetic waves. The radar system operates on radio waves and microwave frequencies. Microwave spectra consist of electromagnetic waves with wavelengths between 1 mm and 1 m (from 300 GHz to 300 MHz) between infrared and radio radiations.

The working principle of the radar system is even simpler than it seems. The basic idea in the radar system is that a signal is sent on the object with the help of the transmitter; this signal hits the object and reflects back. The reflected waves are collected in the receiver so that the object appears on the radar (Richards, Scheer, & Holm, 2010). Here the electromagnetic waves are radio waves and microwaves. The working principle of the Radar system can be seen in Fig. 1.1 After the electromagnetic waves hit the object, some of them are broken and some of them are reflected. The returned reflected waves are detected by the radar system. The object's position, speed, direction and other information are determined by the energy of the reflected waves.

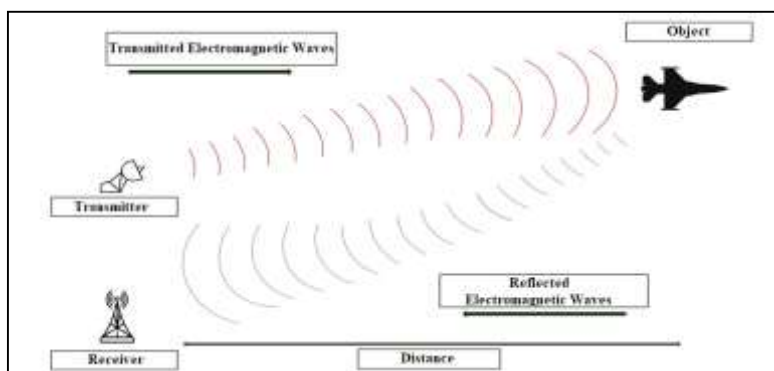


Figure 1.1. The working principle of radar system.

The amount of reflected and refracted signal after being sent onto the object depends on the surface and substance properties. If the material surface consists of a perfectly flat surface, most of the incoming radar signals will be reflected in one direction. However, if the material surface consists of a porous and uneven surface, the incoming radar signals will be refracted in many directions. So this affects the visibility of the object on the radar.

The information obtained from the radar systems is very valuable for the course of military operations and therefore provides a great advantage to the party that has the appropriate radar systems. With the engagement of radar systems, new researches have been started to prevent these systems. In general, the research focused on reducing the scattering directions and amount of electromagnetic waves reflected from the object by absorbing the transmitted waves, and aimed at the appropriate material and design (Kangal, 2013). As a result of these researches, stealth technology or rather low-observable technology, one of the main electronic countermeasures used to make aircraft, ships, helicopters, missiles and other military equipment less detectable, has become an area of interest for many countries (Yuan et al., 2011). The purpose of stealth technology is to reduce the target vehicle's radar cross section (RCS). RCS is a property of the target size, shape and material that it is produced. RCS is a ratio of incoming and reflected power (Saville, 2005). The purpose of reducing the radar cross section is to prevent potential target vehicles from being seen in enemy radar systems.

The radar systems have a stronger structure every day with the development of technology. For this reason, there has been intense interest and increased studies on stealth technology which counter radar systems. Along with these studies, various methods are searched to realize the stealth technology more efficiently. The purpose of these methods is to reduce the reflection of the electromagnetic waves sent and to reduce the radar cross section area.

1.1. Electromagnetic wave basics and absorbing theory

Electromagnetic waves are created by the accelerated motion of electrons or photons. Transverse waves propagating in a perpendicular direction to the plane formed by the electric and magnetic fields, that they are perpendicular each other, are called electromagnetic waves or electromagnetic radiation (Callister & Rethwisch, 2010). Figure 1.2a shows the propagation of generated electromagnetic waves with electric field and magnetic field. Electromagnetic waves are classified in different types according to their frequency or wavelength and they are shown in the electromagnetic spectrum (see Fig. 1.2b). The electromagnetic radiations are

called as Radio Waves, Microwaves, Infrared, Visible Region, Ultraviolet, X-Rays and Gamma Rays with increasing their frequency or decreasing wavelength.

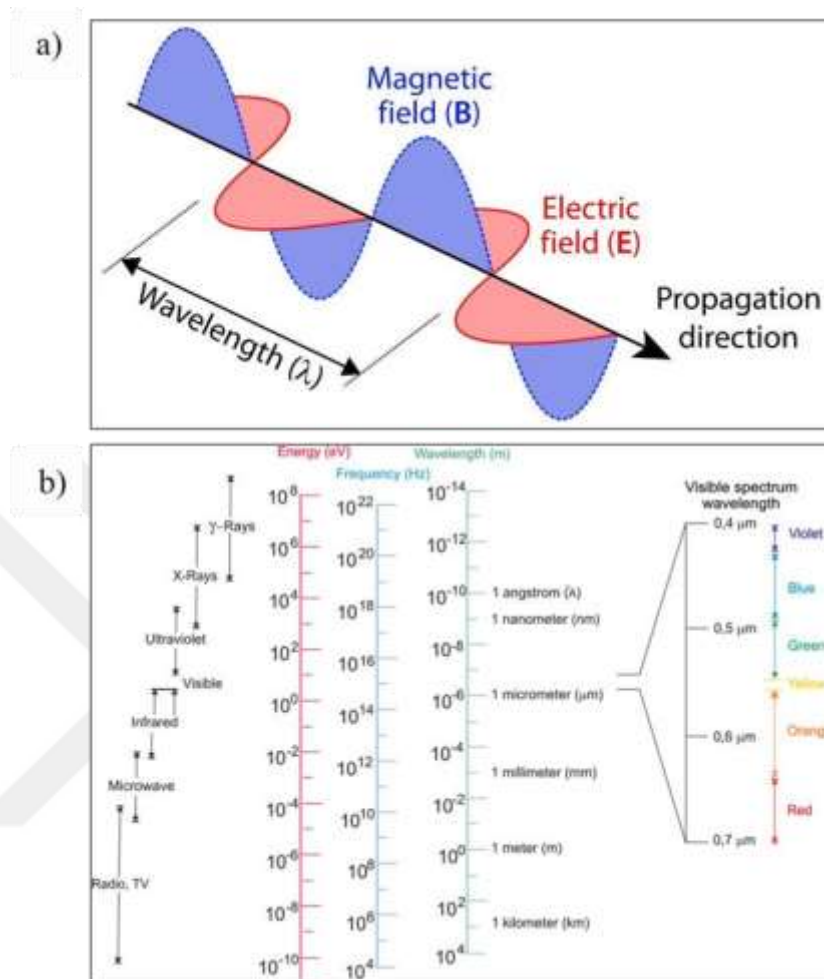


Figure 1.2. a) Creation of electromagnetic waves (Verhoeven, 2017) b) Electromagnetic spectrum (Callister & Rethwisch, 2010).

Microwave spectra in the electromagnetic spectrum consist of electromagnetic waves with wavelengths between 1 mm and 1 m, extending from 300 GHz to 300 MHz between infrared radiation and radio frequencies (Akinay, 2018). Figure 1.3 shows the ranges in microwave spectra with increasing frequency and decreasing wavelength. Additionally, it shows Microwave/Communication bands between 3 GHz and 30 GHz.

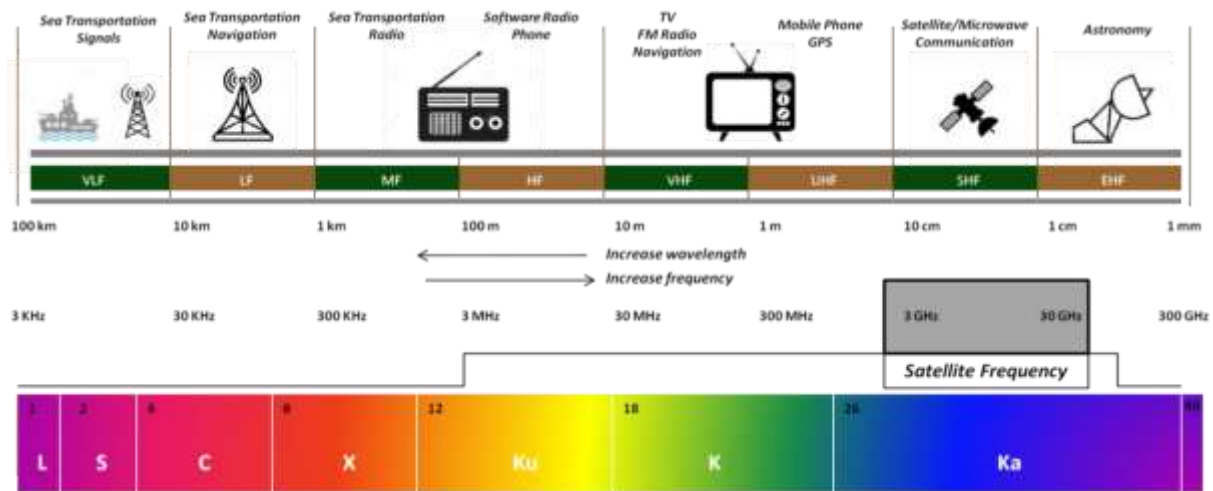


Figure 1.3. Microwave spectrum with increasing frequencies (Hu, 2010).

Detection of objects from nano to macro size is determined by electromagnetic methods. To understand the functions of electromagnetic waves, it is necessary to know the electromagnetic waves and objects interactions. The characters of these interactions vary according to the structure, properties and frequency of the material. Table 1.1 includes the frequency ranges of the radar system and the areas in which it is used within the scope of these frequency ranges.

The propagation of electromagnetic waves are generally defined by permittivity and permeability depending on the frequency (Schick, 2009). In other words, the microwave properties of the material depend on the dielectric and magnetic properties of the material.

Table 1.1. Usage areas of radar frequencies in different bands (Bhatta & Geethapriya, 2016).

BAND DESIGNATION	FREQUENCY RANGE	USAGE AREAS
High Frequency	3-30 MHz	Over The Horizon Surveillance
Very High Frequency	30-300 MHz	Very Long Range Surveillance
Ultra High Frequency	300-1,000 MHz	Very Long Range Surveillance
L	1-2 GHz	Long Range Surveillance En Route Traffic Control
S	2-4 GHz	Moderate Range Surveillance Terminal Traffic Control Long Range Weather
C	4-8 GHz	Long Range Tracking Airborne Weather Detection
X	8-12 GHz	Short Range Tracking Missile Guidance Mapping, Marine RADAR Airborne Intercept
K _u	12-18 GHz	High Resolution Mapping Satellite Altimetry
K	18-24 GHz	Little Use
K _a	27-40 GHz	Very High Resolution Mapping Airport Surveillance

1.1.1. Dielectric mechanism

Permittivity (ϵ) is known as the dielectric constant and is defined as the interaction between the electric field and the material. The real part (ϵ') of the dielectric constant describes how much energy is stored in a material under external electrical field. The imaginary part (ϵ'') of the dielectric constant is known as the loss factor. The complex dielectric constant (ϵ_r) consists of real and imaginary dielectric constants (Arjmand, 2014; Ramajo et al., 2009).

Accordingly, dielectric mechanism, namely permittivity, can be defines as

$$\epsilon = \epsilon' - i \epsilon'' \quad (1)$$

where ϵ' and ϵ'' real and imaginary permittivity, respectively.

1.1.2. Magnetic mechanism

Magnetic permeability (μ) is defined as the interaction of a material with a magnetic field. Complex magnetic permeability (μ_r) also consists of real magnetic permeability (μ') defining energy storage and imaginary magnetic permeability (μ'') defining energy loss (Akinay, 2018). Accordingly, magnetic mechanism, namely permeability, can be defines as

$$\mu = \mu' - i \mu'' \quad (2)$$

where μ' and μ'' real and imaginary permeability, respectively.

1.1.3. Loss tangent ($\tan\delta$)

Another commonly used term is the loss tangent ($\tan\delta$) value that characterizes materials under microwave conditions. This value indicates the material's ability to absorb microwave energy. Dielectric loss factor ($\tan\delta_\epsilon$) and magnetic loss factor ($\tan\delta_\mu$) are two values that contribute to absorbance (Bogdal & Prociak, 2008; Ting & Wu, 2010a). Accordingly, loss tangent can be defines as

$$\tan(\delta_\epsilon) = \frac{\epsilon''}{\epsilon'} \text{ and } \tan(\delta_\mu) = \frac{\mu''}{\mu'} \text{ so } \tan(\delta) = \tan(\delta_\epsilon) + \tan(\delta_\mu) \quad (3)$$

That is called loss tangent because it measures the loss in the medium.

1.1.4. Radar absorbing theory

Radar is a sensitive technological tool for detecting the target object. Radar Absorber Materials are used to hide the detection of objects by protecting them from the electromagnetic radiation emitted by the electronic circuits in the radar systems. Because of this technology, they are very important for modern electronic wars.

In order to understand the radar absorption theory and radar absorber materials, we must first discuss the interaction of electromagnetic radiation with matter. Here, the main purpose is to reduce the reflection coefficient which is the ratio of reflected energy to incoming energy. In attempting to minimize reflection from a surface, it is useful to consider the physical equations that represent the reflection process. There are three conditions that result in minimum reflectivity (Ertuř, 2014).

The first equation explains the reflection coefficient at an interface;

$$r = \frac{\eta_M - \eta_0}{\eta_M + \eta_0} = \frac{Z_M - Z_0}{Z_M + Z_0} \quad (4)$$

where r is the reflection coefficient and η is the admittance of the propagating medium. Admittance is also reverse interaction with intrinsic impedance ($Z = 1/\eta$). Z_M and Z_0 are impedance of media and free space, respectively.

The reflection coefficient falls zero when $Z_M = Z_0$ which means that the material's impedance matches with the incident medium. So incident impedance of free space is given by;

$$Z_0 = \frac{E}{H} = \sqrt{\frac{\mu_0}{\epsilon_0}} \quad (5)$$

where E and H are electric and magnetic field vectors, respectively. μ_0 and ϵ_0 are the permittivity and permeability constants of free space, respectively. Therefore, the material having an impedance of 377 ohms will not reflect electromagnetic waves, if the incident medium is free space (Saville, 2005).

If electric permittivity and magnetic permeability are equal, perfect impedance matching can also be achieved. This result gives the second condition with the minimum reflection coefficient. In this case;

$$\frac{Z_M - Z_0}{Z_M + Z_0} \quad r = \frac{Z_M - Z_0}{Z_M + Z_0} \quad (6)$$

The normalized intrinsic impedance;

$$\frac{Z_M}{Z_0} = \sqrt{\frac{\mu_r}{\epsilon_r}} \quad (7)$$

here;

$$\epsilon_r^* = \frac{\epsilon' - j\epsilon''}{\epsilon_0} \quad \text{and} \quad \mu_r^* = \frac{\mu' - j\mu''}{\mu_0} \quad (8)$$

where prime and double prime of permittivity and permeability terms represents the real and imaginary part, respectively. If the incident medium is free space and the reflectivity is zero,

then μ_r^* becomes equal to ε_r^* . So, the real and imaginary part of the permittivity and permeability are equal, then reflectivity coefficient is zero (Saville, 2005).

The third consideration is the attenuation of the wave as it propagates into the absorbing medium. The power of the wave decays exponentially ($e^{-\alpha x}$) with distance, (x). Here the α is the attenuation constant of the material and can be expressed as (Ertuğ, 2014);

$$\alpha = -\sqrt{\varepsilon_0 \mu_0} (a^2 + b^2)^{1/4} \sin\left(\frac{1}{2} \tan^{-1}\left(-\frac{a}{b}\right)\right) \quad (9)$$

where

$$a = (\varepsilon_r' \mu_r' - \varepsilon_r'' \mu_r'') \text{ and } b = (\varepsilon_r' \mu_r' + \varepsilon_r'' \mu_r'') \quad (10)$$

The α value must be large to achieve a large amount of attenuation with small thickness materials which implies that ε_r' , ε_r'' , μ_r' and μ_r'' . Here, it is notice that there must be large values of permittivity and permeability for this condition to be realized (Saville, 2005).

As a result of these physical equations, absorbers can be characterized by their electrical permittivity and magnetic permeability. In most absorbers, both permittivity and permeability are functions of frequency and can vary significantly even within a small frequency range. If the complex permittivity and permeability are known over a frequency range, the effect of the material on the wave is fully known (Yuzcelik, 2003).

1.2. Radar absorbing structures

Stealth technology is a precaution against radar systems used to make less detectable of aircraft, ships, military vehicles, helicopters, unmanned aerial vehicles and other military technological equipment.

The main purpose of stealth technology is to reduce the radar cross-sectional area (RCS) of the target object and to lose visibility in enemy radar systems. RCS is a property of target size, shape and material from which it is produced, and it is a ratio of incoming and reflected power (Saville, 2005). There are two methods to implement the stealth technology. The first is to reduce the radar cross section area by changing the shape and design of the vehicle and

distributing the incoming electromagnetic waves. Another method is to cover certain parts of the vehicle surfaces with radar absorbing materials (RAM) that absorb electromagnetic waves and lock them into the structure. It is essential to know these phenomena in order to design, manufacture and develop RAM.

1.2.1. Shape of military vehicles

The objective of changing the design and shape of the surfaces and corners of the target vehicle is to change the direction of the incoming electromagnetic waves and thus prevent their reflection. In order to reduce the echo of vehicles against incoming electromagnetic waves, sharp and angled flat surfaces are generally preferred. Figure 1.4a-b show the passenger and fighter aircraft designs, respectively. In Fig. 1.4a, we can see that the plane with a flat surface reflects the radar signals back. So the passenger plane has a high RCS. In Fig. 1.4b, it is seen that the radar cross section area decreases scattered the radar signals with the narrow, sharp and pointed design of the warplane.

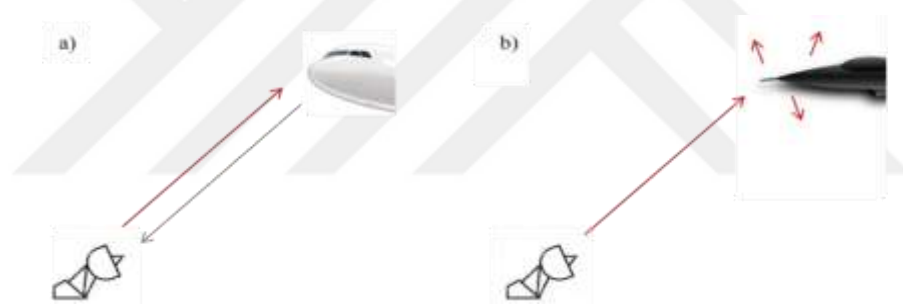


Figure 1.4. Design of a) a passenger plane and b) an aircraft.

It includes some aspects in their designs of modern military warplanes. Examples of military aircraft F-117 Nighthawk, B-2 Spirit bomber aircraft and SR-71 Blackbird are given in the Fig 1.5a-c, respectively.



Figure 1.5. First generation aircrafts with stealth technology. a) F-117 Nighthawk b) B-2 Spirit Bomber and c) Blackbird (Blackbird; Nighthawk; Spirit).

Stealth technology aircrafts are aerodynamically balanced and they achieve high performance and stability through computerized control systems. (Kangal, 2013). Figure 1.6, shows the latest generation warplanes with stealth technology.



Figure 1.6. Latest generation aircrafts with stealth technology. a) Lockheed Martin F-22 Raptor b) Sukhoi Su-57 c) Lockheed Martin F-35 Lightning II (LockheedMartinF-22Raptor; LockheedMartinF-35Lightning-II; SukhoiSu-57).

Vehicle design and shape in stealth technology is also used in sea vehicles. Figure 1.7 shows the stealth technology warships belonging to the naval navies of various countries. Changing the corner angles of warships and designing sharp corner reflectivity decreased the RCS value.



Figure 1.7. Warships with stealth technology. a) Sea Shadow (IX-529) b) HSwMS Helsingborg (K32) c) Kolkata-class destroyer (HSwMSHelsingborg(K32); Kolkataclassdestroyer; SeaShadow(IX-529)).

Vehicle design/shape, which is one of the parameters of privacy technology, is also applied for land vehicles. RCS is reduced by designing the corners, front surface area and rear surface area of land vehicles at certain angles. Figure 1.8 shows that the stealth technology ground war vehicles.



Figure 1.8. Ground war vehicles with stealth technology. a) PL-01 b) Boxer c) Challenger 2 (Boxer; Challenger2; PL-01).

1.2.2. Radar absorbing materials

Radar absorber materials are divided into different groups according to the sources in the literature. The most important properties of these materials are called as permittivity and permeability. Many materials have been used for radar absorption applications; including carbon, metals, ceramics, conductive and insulating polymers (Saville, 2005). The main radar absorber material groups are given as following subtitles.

1.2.2.1. Dielectric absorbers

Dielectric absorber materials do not have the ability to absorb energy alone, due to their low dielectric loss factor. For this reason, composite materials consisting of conductive filling materials together with polymer binder are the most widely used in dielectric absorbers. Conductive fillers are graphite, carbon black, fullerenes, metals, metal oxides and semiconductors (Petrov & Gagulin, 2001). By changing the amount of component materials (dielectric and conductive filler) in the composite, real and imaginary part of permittivity values can be obtained to ensure optimum matching. Materials of this classes offer many advantages over ferrite materials such as they are light as the main material and therefore which do not weigh much in military vehicles (Gaylor, 1989).

1.2.2.2. Magnetic absorbers

The general structure of the ferrite spinel structure is MFe_2O_4 . Here M represents a divalent metal ion ($M=Zn, Fe, Co, Ni, Mn$, etc.). Crystal ferrites have a cubic structure. There are 2 types of ferrite structures such as soft and hard. Soft ferrites can easily change their magnetization when external magnetic field is applied, but hard ferrites can not easily change their magnetization under magnetic field due to their high coercive fields. There has been great interest in soft ferrites as radar absorber materials. Ferrites are ceramic materials. In other words, it shows both very good magnetic and very good mechanical performance at high temperatures up to the Curie points (Gaylor, 1989). For these reasons, ferrites have taken a large place as radar absorbing material in military aircraft. Ferrites are known as the best magnetic components for electromagnetic wave absorbing composites due to their magnetic properties. They are generally used with absorbent rubbers, epoxy resins and various paints as absorbent composite structures in various military vehicles.

1.2.2.3. Oxide absorbers

Oxide-based materials have a wide range of functional capacity of oxygen anion attached to metal-centered atoms to absorb electromagnetic radiation. These types of materials allow the absorbing incoming electromagnetic waves using the dielectric and magnetic loss mechanism and to be used in various potential applications. (Green & Chen, 2019). For this reason, these types of materials have wide range of high electromagnetic absorption potential with magnetic permeability and dielectric permittivity.

1.2.2.4. Polymeric composites

Composite materials are structural materials that combine at least two materials physical and chemical properties, which are insoluble in each other and have different properties before combining. Composite materials consist of matrix as a main phase and reinforcing materials dispersed in matrix. The mechanical properties of the matrix structure are of great importance for the reinforcement elements to exhibit their functions. In radar absorbing composite applications, epoxy resin matrix is generally preferred due to its immense physical and chemical properties such as its easy application, abrasion and corrosion resistance, adhesion strength and heat resistance. With the reinforcement of various magnetic materials (ferrites) in the epoxy matrix, the desired absorption properties are obtained. Magnetic and dielectric reinforcement materials and their concentrations added to epoxy determine the microwave properties of the material. The reinforcement materials added into the polymer matrix materials contribute to the microwave absorption energy depending on the chemical structure and increases the microwave absorption capability of the material.

1.3. Magnetism

Magnetism is the phenomenon of various materials found in nature creating a repulsive or attractive interaction to each other. Materials in nature exhibit different types of magnetic characteristics because the magnetic moments present in their natural structures. Depending on the technology, magnetism and magnetic materials have gained importance for devices such as computers, generators and electric motors. In addition, magnetism has gained importance in the military field with the development of military warfare systems day by day depending on technology. In order to understand the basis and functioning of magnetic materials used in advanced technologies, it is necessary to examine the origin of magnetism.

1.3.1. Origins of magnetism

In the classical atomic model, electrons move around a nucleus with velocity (V) at radius R , that they create a current (I) in the opposite direction to their motion. Accordingly, magnetic moment ($\vec{\mu}$) occurs with this current loop ($\vec{\mu} = I\vec{A}$, here $\vec{A}$ is a normal area vector). The electron-nucleus system is shown schematically in the Figure 1.9.

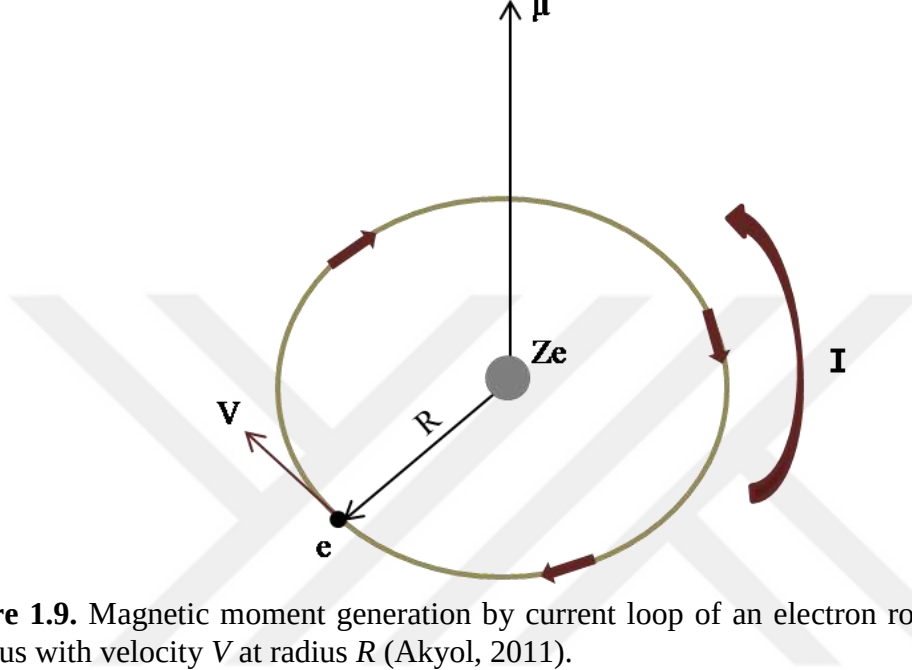


Figure 1.9. Magnetic moment generation by current loop of an electron rotating around the nucleus with velocity V at radius R (Akyol, 2011).

In the electron nucleus phenomenon belonging to the basic hydrogen atom, an electric current is defined as,

$$I = \frac{eV}{2\pi R} \quad (9)$$

where, V and e represents the electron speed and the charge, respectively. For this reason, magnetic moment can be written as,

$$|\vec{\mu}| = |I\vec{A}| = \frac{eV}{2\pi R} \pi R^2 |\hat{A}| \quad (10)$$

The orbital angular momentum of an electron moving around a nucleus can be written as following relation;

$$\vec{L} = m\vec{V} \times \vec{R} \quad (11)$$

When the orbital angular momentum is included in Eq. (10), we can get;

$$|\vec{\mu}| = \frac{1}{2} eVR|\hat{A}| = \frac{e}{2m_e} |\vec{L}| = \gamma |\vec{L}| \quad (12)$$

here γ represents the gyromagnetic ratio. In addition to the equations, electrons belong to a spin angular momentum written by the operator S^2 in addition to the orbital angular momentum.

$$S^2\chi = \hbar^2 s(s+1)\chi \quad (13)$$

In the following equation, the quantized spin angular momentum is given.

$$|\vec{S}| = \hbar \sqrt{s(s+1)} \quad (14)$$

Spin magnetic moment can be expressed as in the following equation,

$$|\mu_s| = \gamma |\vec{S}| = \gamma \hbar \sqrt{s(s+1)} \quad (15)$$

The magnetic moment resulting from the orbital and spin angular momentum mentioned in the above equations can also be written as Bohr magneton ($\mu_B = e\hbar/2m_e$).

$$|\mu_s| = \gamma |\vec{S}| = \mu_B \sqrt{s(s+1)} \quad (16)$$

$$|\mu_l| = \gamma |\vec{L}| = \mu_B \sqrt{l(l+1)} \quad (17)$$

Following the equation shows the total contribution of the orbital and spin angular momentum,

$$|\mu_j| = \gamma |\vec{J}| = \mu_B \sqrt{j(j+1)} \quad (18)$$

here, j is the total angular momentum quantum number that is equal to the sum of the orbital angular momentum and the spin angular momentum.

$$\vec{J} = \vec{L} + \vec{S} \quad (19)$$

1.3.2. Classification of magnetic materials

All of the materials found in nature have different magnetic properties due to their magnetic moment's interactions. As mentioned in the section on the basics of magnetism, magnetism arises from unpaired electrons and natural spins in atoms found in nature. Describing how materials respond to a magnetic field is the best way to understand magnetic material types. There are 5 types of magnetic behaviors of materials in nature which are called as Diamagnetic, Paramagnetic, Ferromagnetic, Antiferromagnetic and Ferrimagnetic.

1.3.2.1. Diamagnetism

Diamagnetic materials do not have a net magnetic moment when no magnetic field is applied. Therefore, when an external magnetic field is applied to such materials, it changes the speed of orbiting electrons that interact with the magnetic field. The direction of magnetic moment in these materials occurs in the opposite direction to the magnetic field, that is, it causes negative magnetic susceptibility behavior by causing the spins of the materials to be reversed. When the applied magnetic field is removed, magnetic susceptibility disappears and magnetization does not occur. Materials exhibit such behavior are classified as 'Diamagnetic'. Water, many plastics, Cu and Al_2O_3 can be given as examples for diamagnetic materials. The magnetic susceptibility of diamagnetic materials is negative and generally very small. Figure 1.10 shows the magnetic moment illustration graph of diamagnetic materials under and without the applied magnetic field.

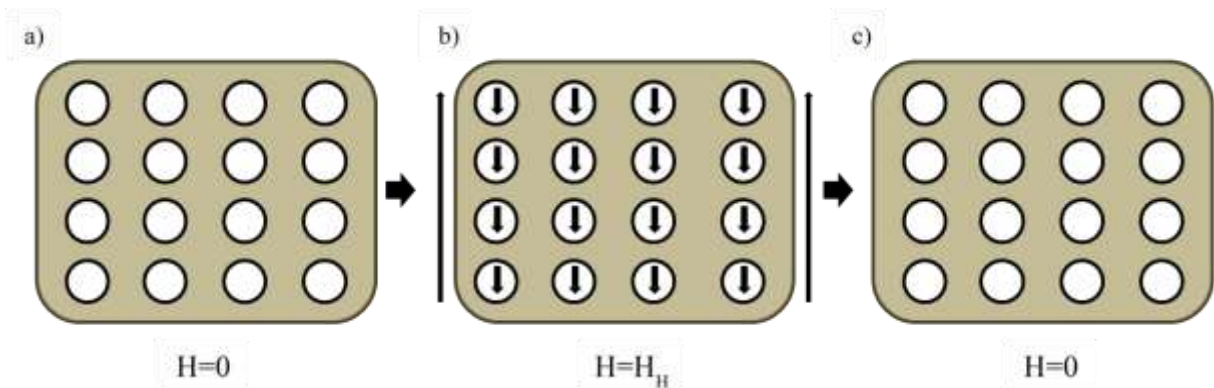


Figure 1.10. Arrangement of magnetic moments of the material exhibiting diamagnetic behavior: a) external magnetic field is not applied, b) external magnetic field is applied and c) external magnetic field is removed.

Figure 1.11 shows the response of the magnetization of material to the applied magnetic field of diamagnetic material. Here, it has been observed that it creates a magnetization in the opposite direction to the field applied on the diamagnetic material. With the increase of the

applied magnetic field, the magnetization of the diamagnetic material also increases in the opposite direction. When the applied magnetic field is removed, the net magnetic moment becomes zero. As a result of these graphs, the magnetic susceptibility of the diamagnetic material is expressed by the equation expressed with $\chi = M/H$ where M is magnetization and H is applied magnetic field. The magnetic susceptibility of diamagnetic materials does not change with temperature (see Fig. 1.11).

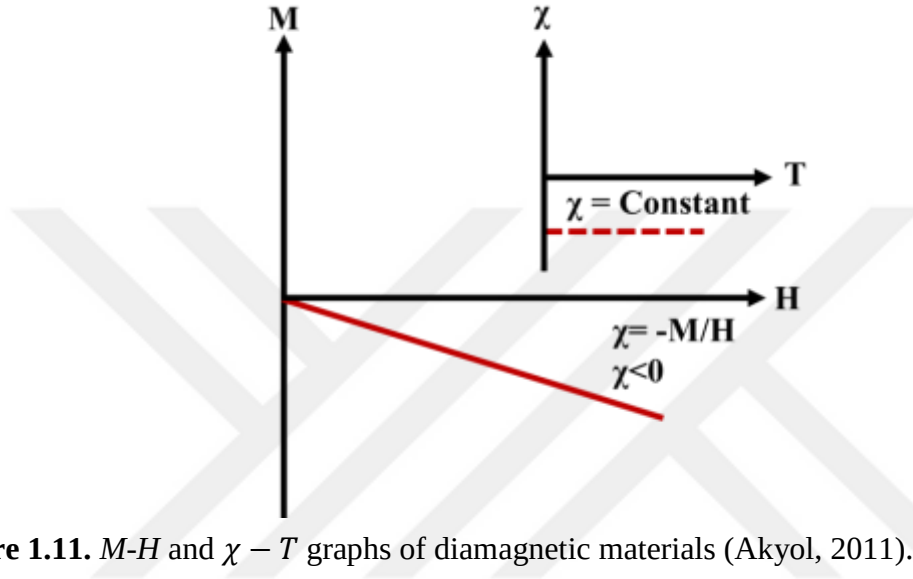


Figure 1.11. M - H and $\chi - T$ graphs of diamagnetic materials (Akyol, 2011).

1.3.2.2. Paramagnetism

Paramagnetic materials are materials that generate a small amount of magnetization in the direction of the applied magnetic field due to the presence of unpaired electrons in their structure. Paramagnetic materials have magnetic moments even though the magnetic field is not applied. However, these magnetic moments are randomly oriented in the structure. For this reason, the net magnetic moment is approximately zero. An external magnetic field is applied on paramagnetic materials, magnetic moments are arranged in the direction of the field. However, the net magnetization value is very small.

As seen in Figure 1.12, when the external magnetic field is removed, the magnetic moments of each atom are randomly oriented. They also have low net magnetization due to their random rotation direction. Aluminium (Al), magnesium (Mg) and tungsten (W) can be given as examples for paramagnetic materials.

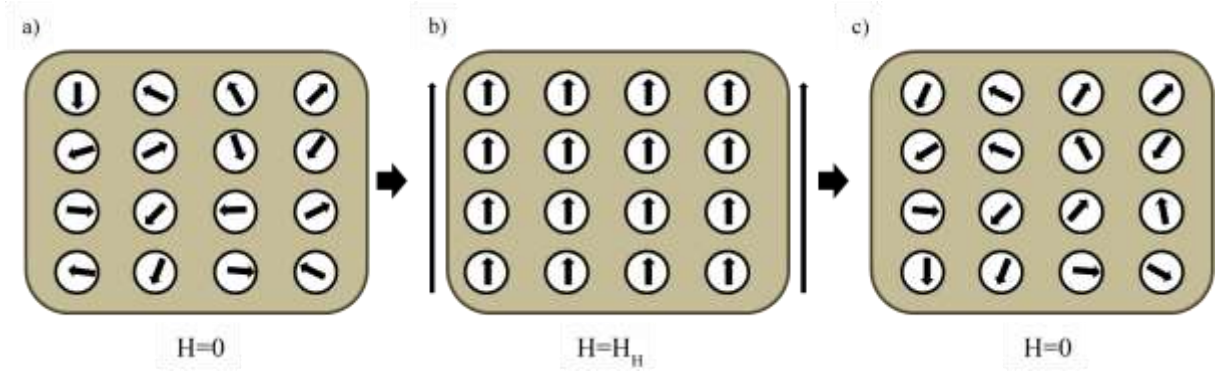


Figure 1.12. Arrangement of magnetic moments of the material exhibiting paramagnetic behavior: a) external magnetic field is not applied, b) external magnetic field is applied and c) external magnetic field is removed.

Figure 1.13 shows the magnetic response of the paramagnetic material against the applied magnetic field, as well as it shows magnetic susceptibility as a function of temperature. With the application of an external magnetic field to the material, the magnetic moments arranged irregularly in the material started to be oriented with the increasing field in the same direction. The total magnetization of the material tends to increase linearly when the external magnetic field applied on the material. As the applied field increases, the arrangement of magnetic moments in the same direction increases. In opposite to the diamagnetic materials, the susceptibility depends on the temperature.

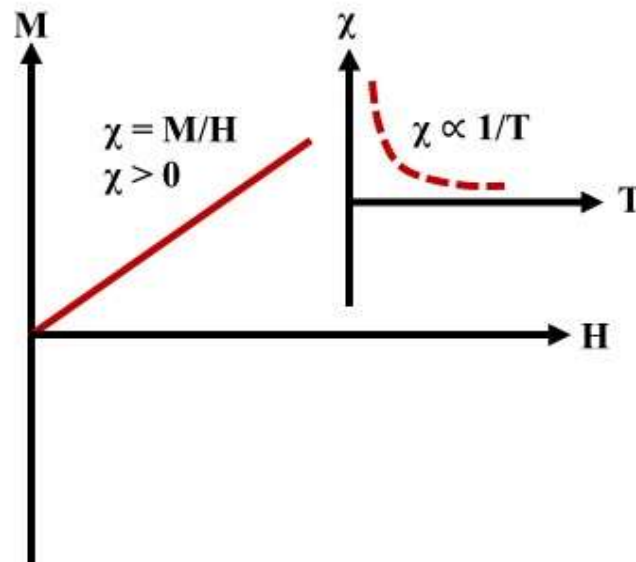


Figure 1.13. M - H and $\chi - T$ graphs of paramagnetic materials (Akyol, 2011).

1.3.2.3. Ferromagnetism

In ferromagnetic materials, the spin bonding formed by unpaired electrons creates magnetic moment patterns of the atoms. All ferromagnetic materials exhibit variable magnetic behavior, ie losing their ferromagnetic behavior, at a certain temperature value due to thermal fluctuations. This temperature is called the Curie temperature (T_C). Ferromagnetic materials are systems with long-distance order at the atomic scale because they have unpaired electron spins arranged parallel to each other in regions known as domains (Akyol, 2011). In short, a ferromagnetic material has a permanent magnetic property even in the absence of an external magnetic field.

Materials that exhibit such behavior are classified as 'Ferromagnetic' which materials such as iron (Fe), nickel (Ni), cobalt (Co) and some rare earths (Gd, ie) are examples of this class.

Figure 1.14a-b show the behavior of magnetic moments when an external magnetic field is not applied and applied on the ferromagnetic material, respectively. As shown in Figure 1.14a, the materials are formed by combining many domains. Each domain has its own net magnetic moment. In Figure 1.14b, as a result of the external magnetic field applied on the ferromagnetic material, the magnetic moments in the domain are arranged according to this field and its magnetism increases in the direction. This magnetization is much greater than paramagnetic materials (Akyol, 2011).

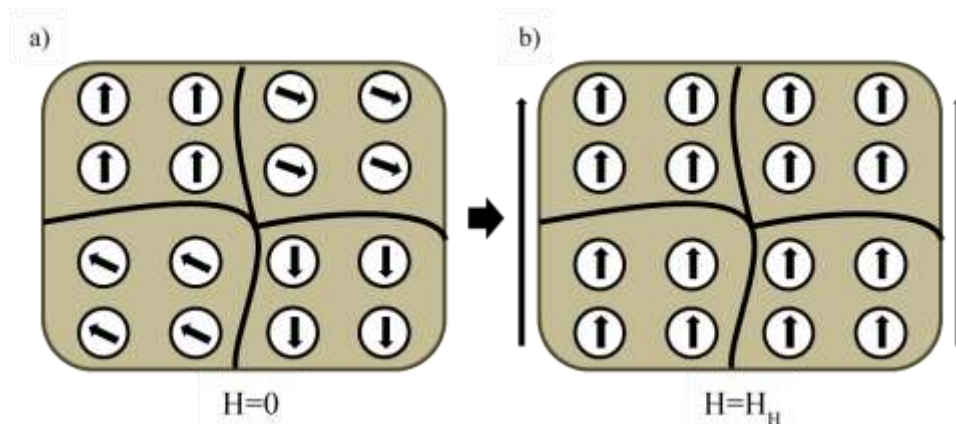


Figure 1.14. Arrangement of magnetic moments of the material exhibiting ferromagnetic behavior: a) external magnetic field is not applied and b) external magnetic field is applied.

Ferromagnetic materials exhibit an irreversible and non-linear behavior under applied magnetic field. This behavior of the ferromagnetic group is explained by the hysteresis loop. Figure 1.15a shows the magnetization response of the ferromagnetic materials against the applied magnetic

field. Figure 1.15b-c show the characteristic hysteresis curves of hard and soft ferromagnetic materials against externally applied magnetic field (Coey, 2010).

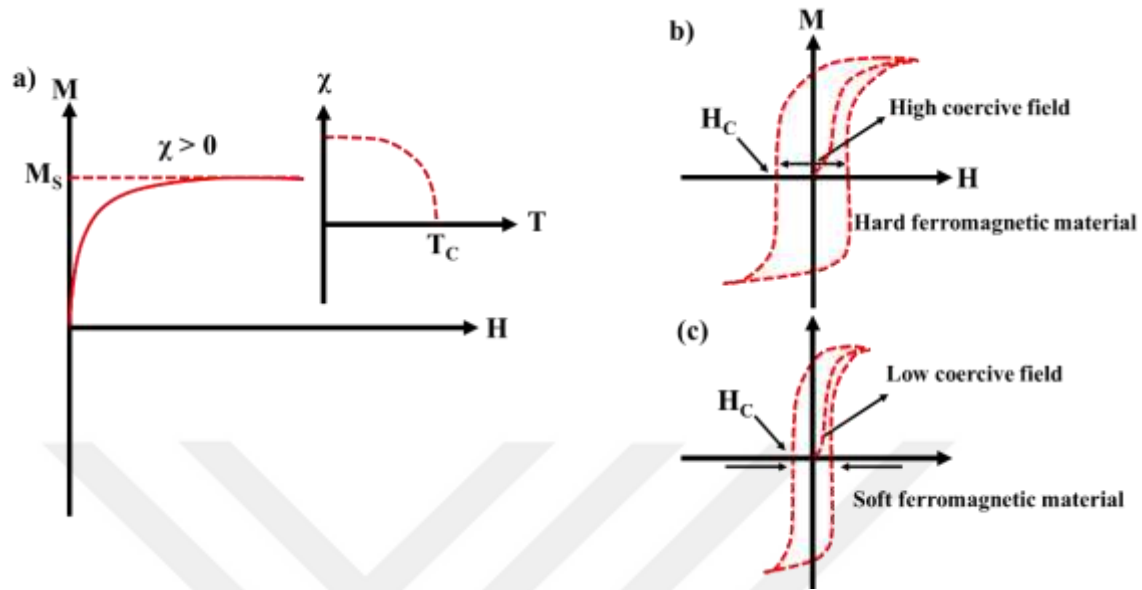


Figure 1.15. a) M - H and $\chi - T$ graphs of ferromagnetic materials. Hysteresis curves of b) hard and c) soft ferromagnetic materials (Akyol, 2011).

As can be seen from Figure 1.15a, as a result of the external magnetic field applied on the material, the atoms in the structure begin to align and orient in the field direction. Magnetization of the material increases in a short time. Magnetization of the ferromagnetic material reaches the saturation magnetization value (M_s) with the increasing applied field (Coey, 2010). Figure 1.15b shows that hard ferromagnetic materials have a large coercive field (H_C). When enough field is applied to these materials, the materials become saturated and their magnetization becomes permanent. Soft ferromagnetic materials appear to have a small coercive field at Figure 1.15c.

1.3.2.4. Antiferromagnetism

The atoms in the antiferromagnetic materials have a net magnetic moment. But the magnetic moments in atoms are in an array that is not parallel to each other, and the closest magnetic moments are dynamics that can cancel each other out. The anti-parallel magnetic moment orientations of an antiferromagnetic material is due to the superexchange interaction (Coey, 2010). Antiferromagnetic materials have a small positive magnetic susceptibility because they have anti-parallel array. Figure 1.16a-b show the behavior of magnetic moments when an external magnetic field is not applied on the antiferromagnetic material and when it is applied,

respectively. Figure 1.16a shows the magnetic moments of neighboring atoms that destroy each other, under zero magnetic fields. In Figure 1.16b, in the same direction the applied field.

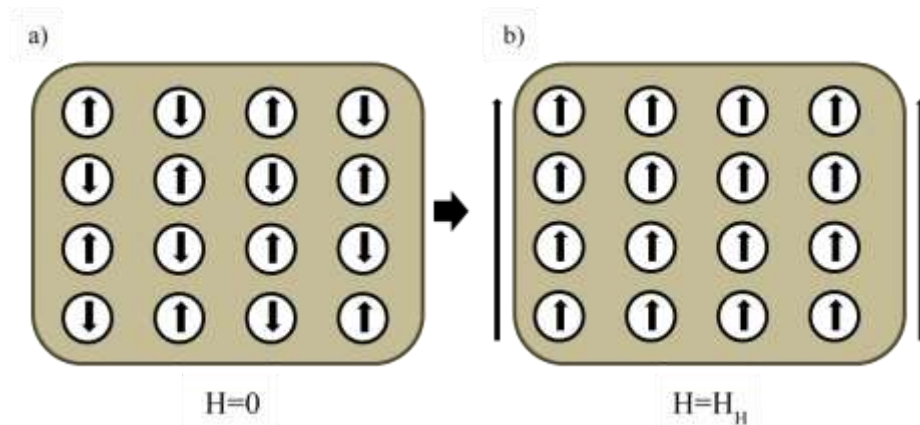


Figure 1.16. Arrangement of magnetic moments of the material exhibiting antiferromagnetic behavior: a) external magnetic field is not applied and b) external magnetic field is applied.

Figure 1.17 shows the M - H graph of antiferromagnetic materials. As a result of the magnetic field applied on the antiferromagnetic material, magnetic moments with different orientations are arranged in the direction of the applied field. It can be observed that the magnetization increases with increasing applied magnetic field on the material. When the magnitude of the magnetic field is increased, if it reaches a sufficient level, the material reaches the magnetization saturation value.

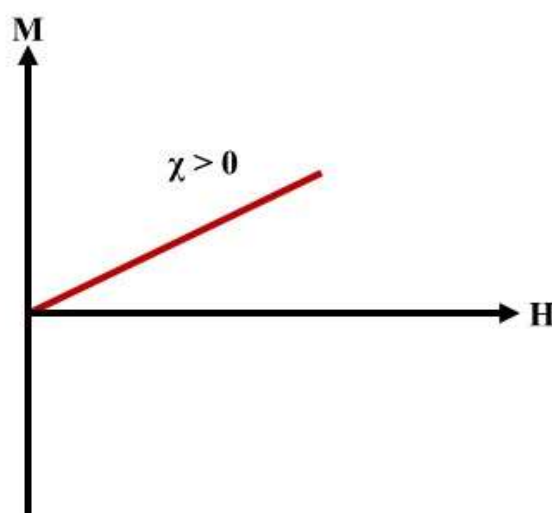


Figure 1.17. M - H graph of ferromagnetic materials (Akyol, 2011).

1.3.2.5. Ferrimagnetism

Ferrimagnetic materials show similarity to antiferromagnetic material structure in terms of magnetic moment sequences of atoms in the structure. But the magnitudes of magnetic moments in atoms are different. Magnetic moments of different magnitude do not severely destroy each other as in antiferromagnetic structure. For this reason, they have a net magnetization. In terms of hysteresis behavior, it behaves similar to the ferromagnetic materials. But the saturation magnetization value of ferromagnetic materials is higher than the ferrimagnetic materials. Figure 1.18a-b show the behavior of magnetic moments when an external magnetic field is not applied on the ferrimagnetic material and when it is applied, respectively.

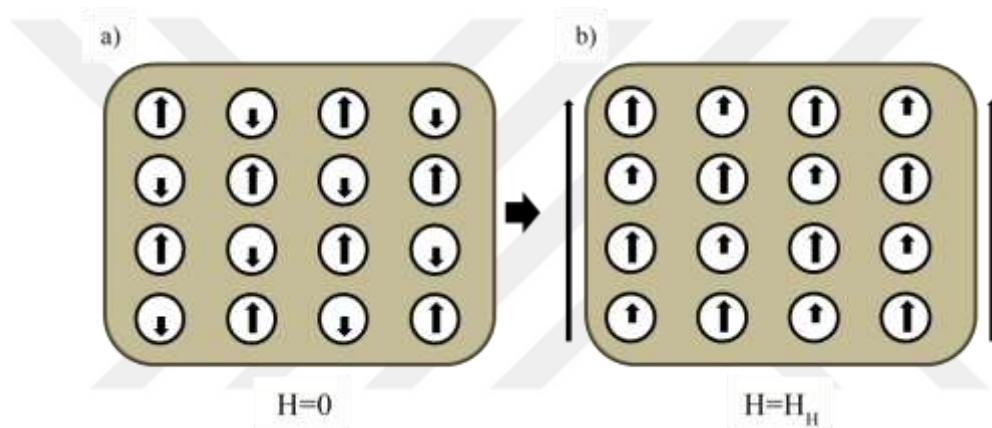


Figure 1.18. Arrangement of magnetic moments of the material exhibiting ferrimagnetic behavior: a) external magnetic field is not applied and b) external magnetic field is applied.

Figure 1.19 shows the M - H graph of ferrimagnetic materials. The response of the ferrimagnetic material to the external magnetic field applied on it is similar to that of the ferromagnetic material. The magnetic moments of the atoms direct in the same direction with the applied magnetic field. The tendency of the magnetic moments to align in that direction increases more and the material reaches the magnetic saturation value when sufficient magnetic field is applied. The saturation magnetization value of ferrimagnetic materials is lower than ferromagnetic materials.

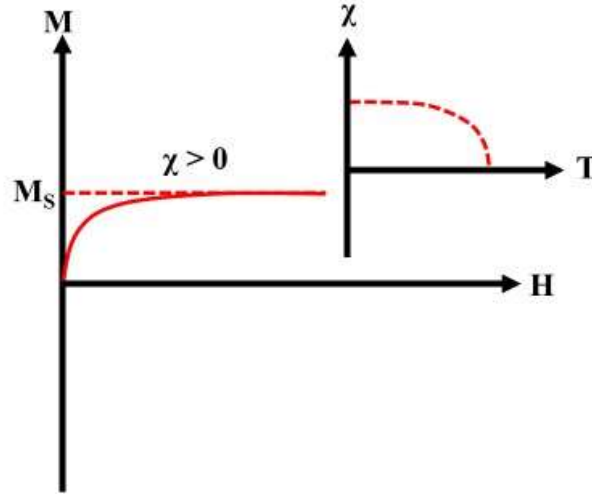


Figure 1.19. M - H and $\chi - T$ graph of ferrimagnetic materials (Akyol, 2011).

1.3.3. Curie temperature and Curie-Weiss law

The critical temperature at which ferromagnetic materials pass into the paramagnetic behavior state is called the Curie temperature. Magnetic dipoles in the ferromagnetic material deteriorate and exhibit paramagnetic behavior with the increasing of temperature. The magnetic susceptibility of the material is the way it responds to the applied magnetic field and is expressed by the following equation.

$$\chi = \frac{M}{H} = \frac{C}{T} \quad (20)$$

Here, C represents the Curie constant. Magnetization is expressed as magnetic moment per unit volume or mass. It also depends on the interactions of the magnetic moments of the atoms in the material. The transition of ferromagnetic to paramagnetic behavior at this critical Curie temperature is expressed as follows.

$$\chi = \frac{C}{T - \theta} \quad (21)$$

The expressed in this mathematical equation is referred to as the Curie-Weiss Rule (Weiss, 1906). The θ symbol represents the temperature constant. It can get negative, zero and positive values in antiferromagnetic, paramagnetic and ferri-/ferro-magnetic materials, respectively.

- If θ equals zero, Curie's law is in effect (Equation 20).
- If θ is positive, the material is in ferri-/ferro-magnetic behavior below the Curie critical temperature.

- If θ is negative, the material has antiferromagnetic behavior below Neel critical temperature (T_N) .

The critical Neel and Curie transition temperatures are shown in Figure 1.20. The critical Curie temperature mentioned here is a characteristic for materials exhibiting ferromagnetic behavior. Neel temperature is a characteristic for materials exhibiting antiferromagnetic behavior (Gülkesen, 2020). The material which is the ferromagnetic (antiferromagnetic) exhibits paramagnetic property above the Curie (Neel) temperature (Coey, 2010).

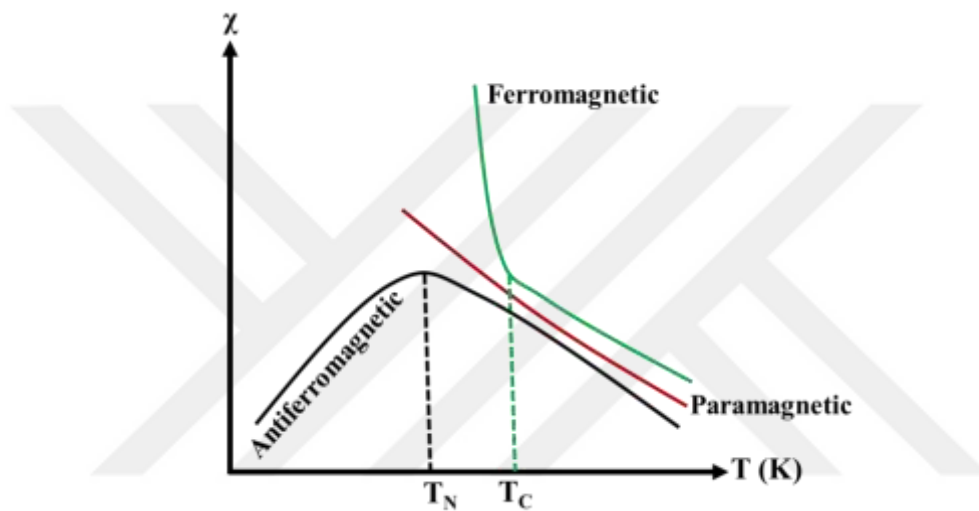


Figure 1.20. Magnetic susceptibility curves of ferromagnetic, antiferromagnetic and paramagnetic materials (Akyol, 2011).

1.3.4. Magnetic hysteresis loop

Hysteresis curves are characteristic curves that show the complex change in magnetization of ferro and ferrimagnetic materials by applied magnetic fields. As a result of these curves, a lot of information can be learned about the magnetic characteristics of materials. Saturation magnetization (M_s), remenant magnetization (M_r) and coercive field (H_c) are obtained from the hysteresis loop. In addition to these, information about the magnetic softness or hardness of the material is obtained. Figure 1.21 shows the hysteresis loop of a multi-domain magnetic material.

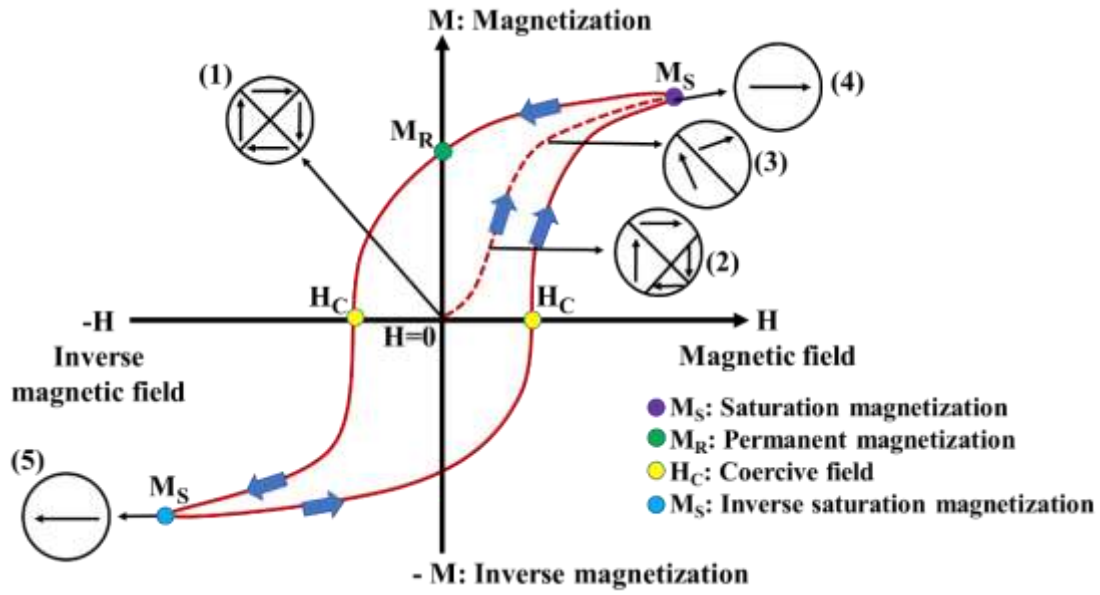


Figure 1.21. Hysteresis loop of a multi-domain magnetic material (Akyol, 2011).

Even though no external magnetic field is applied, ferromagnetic and ferrimagnetic materials have a net magnetization value. Their magnetization increases significantly and reaches a saturation value rapidly (M_s) under applied magnetic field. When the external magnetic field is removed, most of the material shows remanent magnetization (M_r). H_c determines whether the materials are magnetically hard or soft material.

1.4. Thesis objectives

In this thesis, first of all, it is aimed to produce $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ ($0.0 \leq x \leq 1.0$, with 0.25) and Ni_yZnO ($y=0$ and ≤ 0.1) nanoparticles by sol-gel method. The structural and magnetic properties of these nanoparticles will be characterized by performing x-ray diffractometer, scanning electron microscope, energy-dispersive x-ray spectrum and vibrating sample magnetometer to determine the crystal structure, phase determination, particle size, elemental analysis and magnetic properties of the produced nanoparticles. Secondly, it is aimed to produce various advanced radar absorbing composite structures of 4 mm thickness by reinforcing the appropriate amount of nanoparticles into an epoxy matrix. It is aimed to enhance the reflection loss and absorption bandwidth in order to increase the potential application of the produced composite materials in various military vehicles. For this reason, it is aimed to investigate the electromagnetic properties in the X-band and mechanical properties (hardness and impact) of the produced radar absorber composite materials. It is foreseen that radar absorber composite structures will be developed to use in military air, sea and land systems.

2. LITERATURE REVIEW

In this part of the thesis, studies on the production of various radar absorbing materials and composites and their characterization are discussed. We will provide a brief summary of the literature works on radar absorbing systems.

Ting et. al. (2010) worked on the production of Polianilin/BaFe₁₂O₁₉ (PANI/Ba-ferrite) composites by in-situ polymerization method at different aniline/Ba-ferrite weight ratios (Ani/Ba-ferrite = 1/2, 1/1 and 2/1). The Fourier-Transform Infrared spectroscopy (FTIR) analysis showed that Ba-ferrite made a tight bond with PANI. Also, it was observed that only Ba-ferrite pure crystal was present in X-Ray Diffraction (XRD). Ultra-Violet Visible (UVVIS)

and Electron Spin Resonance (ESR) measurements showed the electrical conductivity and interaction between Ba-ferrite and PANI. It showed good absorption performances with good electromagnetic properties in the range of 2-18 and 18-40 GHz. But, epoxy-PANI/Ba ferrite composite was showed better absorption bands at 2–18 GHz than epoxy-Ba ferrite composite. The reflection loss of the epoxy/Ba-ferrite composite sample was observed as -12 dB at 32.3 GHz. Reflection loss values of the epoxy-PANI/Ba ferrite composite samples were obtained as -10.5 dB, -11 dB and -12 dB at 28, 24 and 21.5 GHz, respectively (Ting & Wu, 2010b).

Gama et al. (2011) worked on analysis of measurements of the complex magnetic permeability (μ_r), dielectric permittivity (ϵ_r) and production of radar absorbing composite materials which were formed by MnZn ferrite particles and silicone rubber. XRD analysis showed that presence of single cubic phase and crystal structure of sample were in good agreement for $\text{Mn}_{0.66}\text{Zn}_{0.34}\text{Fe}_2\text{O}_4$. In both cases of composite material samples, complex permeability and complex permittivity were showed a general dependence on the volume fraction of the filler MnZn ferrite at 2-18 GHz. In the increased MnZn ferrite volume fraction, reflection loss (RL) was observed to increase from -5 dB for $v_f=0.10$ to -35 dB for $v_f=0.20$. Here, v_f is the MnZn ferrite fraction. The obtained parameters indicated that samples with higher thicknesses showed better absorption properties. It was also found that for higher MnZn ferrite fraction, smaller thicknesses could offer better absorption properties for higher frequency bands (≥ 10 GHz) (Gama, Rezende, & Dantas, 2011).

Bayrakdar (2011) worked on investigation of electromagnetic properties of the nanoparticles/paraffin nanocomposite materials of $\text{Co}_x\text{Mn}_{1-x}\text{Fe}_2\text{O}_4$ spinel ferrite (where $x=0.0, 0.5$ and 1.0) at 8-20 GHz. $\text{Co}_x\text{Mn}_{1-x}\text{Fe}_2\text{O}_4$ nanoparticles were synthesized by cetyltrimethylammonium assisted hydrothermal route using NaOH. They work on the phase purity and crystallinity of the nanoparticles by performing the powder XRD method. The crystal structure of nanoparticles are formed as a spinel cubic structure. The grain sizes of spinel structures are 62 nm, 60 nm and 58 nm for $x=0.0, 0.5$ and 1.0 in $\text{Co}_x\text{Mn}_{1-x}\text{Fe}_2\text{O}_4$ spinel ferrite, respectively. A reflection loss of -46.60 dB at 10.5 GHz with a bandwidth approximately 2 GHz in a sample of 2 mm absorber composite thickness was observed (Bayrakdar, 2011b).

M. Wang et al. (2015) worked on conductive polyaniline (PANI) and nickel zinc ferrite binary composites synthesized by in-situ polymerization process. The electromagnetic absorption properties of the composites were also investigated. According to XRD curves, pure ferrite and

its composites at different molar ratios had diffraction peaks for spinel type crystals. FTIR spectra showed the PANI (1562, 1481, 1301, 1109, and 799 cm^{-1}) peaks and NiZn ferrite (579 and 390 cm^{-1}) bonds which indicate the presence of both NiZn ferrite particles and PANI. Thermogravimetry (TG) curves of pure PANI and composite materials with different molar ratios showed the amount of ferrite in the composite. It was also found that the best electromagnetic absorption property and wide absorption bandwidth would be obtained by $n(\text{PANI}):n(\text{Ni}_{0.6}\text{Zn}_{0.4}\text{Fe}_2\text{O}_4)=1:2$ ratio. The highest reflection loss of $\text{Ni}_{0.6}\text{Zn}_{0.4}\text{Fe}_2\text{O}_4/\text{PANI}$ in the ratio of 1:2 was achieved as -41 dB at 12.8 GHz, and the bandwidth at -10 dB was achieved 5 GHz with 2.6 mm absorber thickness (M. Wang et al., 2015).

G. Wang et al. (2015) worked on the production of the ZnO coated by reduction of NiO nanoparticles with an Atomic Layer Deposition (ALD) method. XRD results showed that all diffraction peaks for ZnO index to the hexagonal phase. The Scanning Electron Microscope (SEM) and Transmission Electron Microscope (TEM) images showed that the surfaces of ZnO were densely coated with Ni nanoparticles with narrow particle size distribution and average size of Ni nanoparticles were found as 13.1 nm. The optimal RL calculated from the measured complex permittivity and permeability was found as -48.0 dB at 10.4 GHz. Furthermore, the electromagnetic wave absorption less than -10 dB was found to reach 5.3 GHz for 1.5 mm thickness sample. It has been seen in microwave analysis that ZnO@Ni exhibits significantly improved electromagnetic absorption properties compared to ZnO (G.

Wang et al., 2015).

Ma et. al. (2017) worked on the preparation of spinel cobalt chrome zinc ferrites ($\text{Co}_{0.7}\text{Cr}_{0.1}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$) and polyaniline (PANI)- $\text{Co}_{0.7}\text{Cr}_{0.1}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ composites by polyacrylamide gel and in-situ polymerization method. As a result of the structural analysis, (PANI)- $\text{Co}_{0.7}\text{Cr}_{0.1}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$, $\text{Co}_{0.7}\text{Cr}_{0.1}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ and $\text{Co}_{0.8}\text{Cr}_{0.2}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ were successfully produced and no impurities were observed. But, since Co^{2+} is replaced by Cr^{3+} in octahedron ferrites, the lattice parameter of $\text{Co}_{0.8}\text{Cr}_{0.2}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ ferrites drops from 0.8409 to 0.8377 nm. The values of saturation magnetization (M_s), remenant magnetization (M_r) and coercive field (H_c) for (PANI)- $\text{Co}_{0.7}\text{Cr}_{0.1}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ composites are $8.80 \text{ mA.m}^2.\text{g}^{-1}$, $3.14 \text{ mA.m}^2.\text{g}^{-1}$ and 37.22 kA.m^{-1} , respectively, which are less than those of the $\text{Co}_{0.7}\text{Cr}_{0.1}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ ferrites. In the 5–20 GHz frequency band range, the maximum values of the two reflection losses of 13.17 dB and -

15.36 dB of (PANI)-Co_{0.7}Cr_{0.1}Zn_{0.2}Fe₂O₄ composites appear at 14.1 and 17.9 GHz, respectively (Ma, Shang, Wang, & Jiang, 2017).

Handoko et al. (2017) worked on the synthesis of the BaFe_{12-2x}(Co_{0.8}Ti_{0.8}Mn_{0.4})_xO₁₉ ($x=0$ and $x=1$) by a ceramic method. The ferrite powders which processed at 1100 °C for 5 hours, were mixed with resin for the production of composite material in 2 mm thickness. According to the XRD results, BaFe_{12-2x}(Co_{0.8}Ti_{0.8}Mn_{0.4})_xO₁₉ samples ($x=0$ and $x=1$) showed that they had no impurities and contamination. It was observed that single phase with a hexagonal structure. The samples in larger coercivity at $x=0$ and small coercivity at $x=1$ had higher permeability and wide microwave absorption ability. Co, Ti and Mn elements substituted samples showed interesting reflection losses in the between 8.2-12.4 GHz (Handoko et al., 2017).

Yin et. al. (2018) worked on the production of ZnO/Fe₃O₄/paraffin composites prepared by physical blending technology. According to the XRD diffraction results, ZnO and Fe₃O₄ diffraction peaks were good agreement with the literature. ZnO micro powder added to the structure contributed to microwave absorption by increasing the dielectric loss and magnetic loss of the composite. The mass ratios of ZnO and Fe₃O₄ were used as 1:2 and 1:4, respectively. The absorption bandwidth of 0.93 GHz (1.59 GHz to 2.52 GHz) and 0.85 GHz (1.26 GHz to 2.11 GHz) measured at -10 dB was obtained in the frequency range of 0.5-3 GHz. Materials in this structure are used as an effective and efficient microwave absorber in applications to be used in accordance with low frequency ranges such as 0.5-3 GHz (Yin et al., 2018).

Rehman et al. (2019) worked on production the composite of polyaniline (PANI) with three dimensional (3D) ZnO microspheres synthesized by single pot solvothermal and in-situ polymerization method. It was observed that the XRD result obtained as a result of the structural analysis matched with the literature and no impurities were seen. ZnO@PANI composite has been observed to increase complex permeability and dielectric loss at 2-18 GHz frequency range. It has been found that the maximum reflection loss value of ZnO@PANI reaches -41 dB at 14 GHz. The material having 3.5 mm thickness is superior to its composite with materials of different thicknesses (ur Rehman, Liu, Ahmed, & Bi, 2019).

Vishal et. al. worked on the production of Co_xNi_{1-x}Fe₂O₄ ($x=0$ and $x=0.4$)/carbon nano fiber reinforced epoxy matrix nanocomposites for radar absorbing structures. Co_xNi_{1-x}Fe₂O₄ ($x=0$

and $x=0.4$) samples were synthesized by heat treatment method. According to the XRD results of the produced nanoparticles, the characteristic diffraction peaks were formed, but only one α - Fe_2O_3 impurity peak appeared. The produced $\text{Co}_x\text{Ni}_{1-x}\text{Fe}_2\text{O}_4$ nanoparticles were first dispersed 45% into the epoxy matrix. Microwave measurement results showed an absorption (NC_4 containing 4% Co) under -10 dB at 1.4 GHz (10.3 mm thickness). Secondly, ferrite/epoxy systems were prepared by adding 1%, 2% and 3% carbon nano fiber. Figure 2.1 shows a reflection loss vs frequency of nanocomposite structures in various structures. Optimum value was obtained in NC_4CF_2 structure with 1.6 mm thickness and 3.23 GHz absorption bandwidth. These structures are potential for radar applications, aviation applications and privacy technologies (Kumar Chakradhary & Akhtar, 2021).

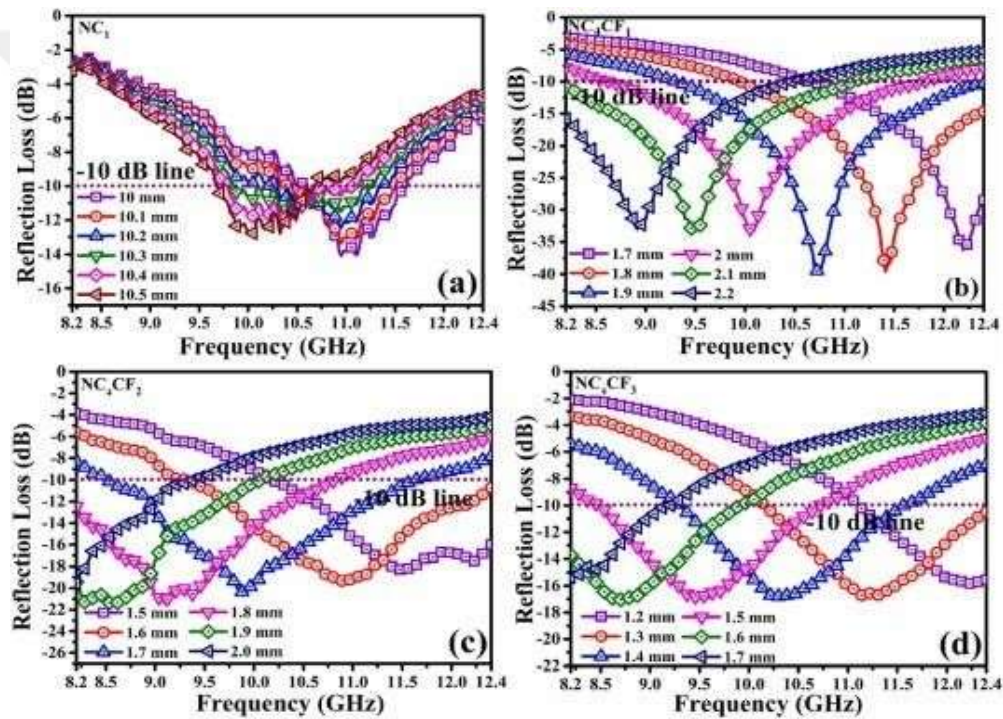


Figure 2.1. Reflection loss versus frequency graph of nanocomposites (Kumar Chakradhary & Akhtar, 2021).

3. MATERIALS AND METHODS

There are two nanomaterials production types which are called as top-down and bottom-up productions (Hakan Ateş, 2015). In the top-down production method, the materials are processed as a whole and then cut into small pieces on a nanometer scale. Lithography, thermal method, etching, gas atomizer techniques can be given as examples to this group. Another method is called as bottom-up production. Here, the structure is obtained as a result of the growth of atoms or chemical reactions. Sol-gel, flame synthesis, ultrasonic spray pyrolysis, atomic layer precipitation, chemical vapor condensation, wet chemical synthesis techniques can be given as examples of this group. Among the different production methods, the sol-gel method is advantageous due to its low cost. In addition to the cost, it is possible to produce high purity and homogeneity particles at low synthesis temperature by this method.

In this section, the preparation procedures of Ni_yZnO ($y=0.0$ and 10.0 %) and $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ ($0.0 \leq x \leq 1.0$, with 0.25 increments) nanomaterials and radar absorbing composite structures will be explained. The characterization techniques for the investigation of structural and magnetic properties of nanoparticles will be given in this section. In addition, the necessary measurement systems to investigate the electromagnetic and mechanical properties of nanoparticles reinforced epoxy composite structures will be given in detail.

3.1. Sol-gel process

Sol-gel is a method which is obtained by molecular control of the initial organic and inorganic compounds such as pre-initiators (metal salts and metal alkoxides), catalyst and solvent in appropriate amounts. In this method, materials enables high purity, uniform crystal morphology

and homogeneous nanoparticle synthesis with the dissolution of chemical materials in the form of a solution on a molecular basis in the reaction path in liquid (Gülkesen, 2020). The sol-gel process involves the transition of sol composition from a liquid "sol" phase to a solid "gel" phase (Gupta & Chaudhury, 2007). This method, as its name suggests, means solution-gelation. The first stage is the formation of colloidal suspension solution of solid particles in the liquid (Siouffi, 2003). The second part of the process is gelification that it occurs when the viscosity of sol particles decreases with hydrolysis and condensation. Figure 3.1 shows the scheme of the general sol-gel process. After the left colloids are formed, gelation occurs. Here, the left particles do not collapse due to the intermolecular attraction force.

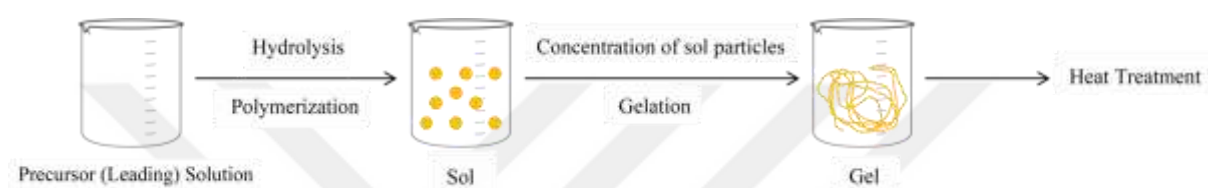


Figure 3.1. Operation scheme of the general sol-gel method.

Sol-gel method is preferred for many reasons. The necessary situations for the production of the material in working conditions can be easily provided. Preparing the pre-starting materials at low temperatures eliminates the risk of thermal degradation and ensures that materials with high purity and homogeneity are produced. The shape and dimensions of the produced material can be easily controlled. For these and many other reasons, the sol-gel method is a preferred and widely used method.

3.2. Preparation of nanomaterials

3.2.1. Synthesis of $\text{Ni}_y\text{:ZnO}$ nanoparticles

There are various production methods for making desired products with different particles in shape, size and structure of ZnO which are physical deposition, precipitation in water solution, hydrothermal synthesis, sol-gel process, precipitation from micro emulsions and mechano-chemical process. (Kołodziejczak-Radzimska & Jesionowski, 2014). In this thesis study, $\text{Ni}_y\text{:ZnO}$ nanoparticles were synthesized by sol-gel method due to its reduced cost, low synthesis temperature, high purity and homogeneity. Here, the Ni ratio determined as 0% and 10%. In this method, 'Nickel Nitrate Hexahydrate- $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ', 'Zinc Nitrate Hexahydrate- $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ' and 'Citric Acid' were used as starting materials. The starting materials (zinc nitrate, nickel nitrate and citric acid) dissolved appropriate stoichiometric proportions in pure

water poured into an empty baker that it was placed on the magnetic stirrer and heater at a temperature of 50-60 °C. The heated magnetic stirrer was set to 200-300 °C and the appropriate amount of gelling agent (ethylene glycol) was slowly added to the solution. After the reaction took place and gelation was achieved, the production process was completed. The produced Ni_yZnO gel was ready for heat treatment. Figure 3.2 shows the solgel production scheme of Ni_yZnO nanoparticles.

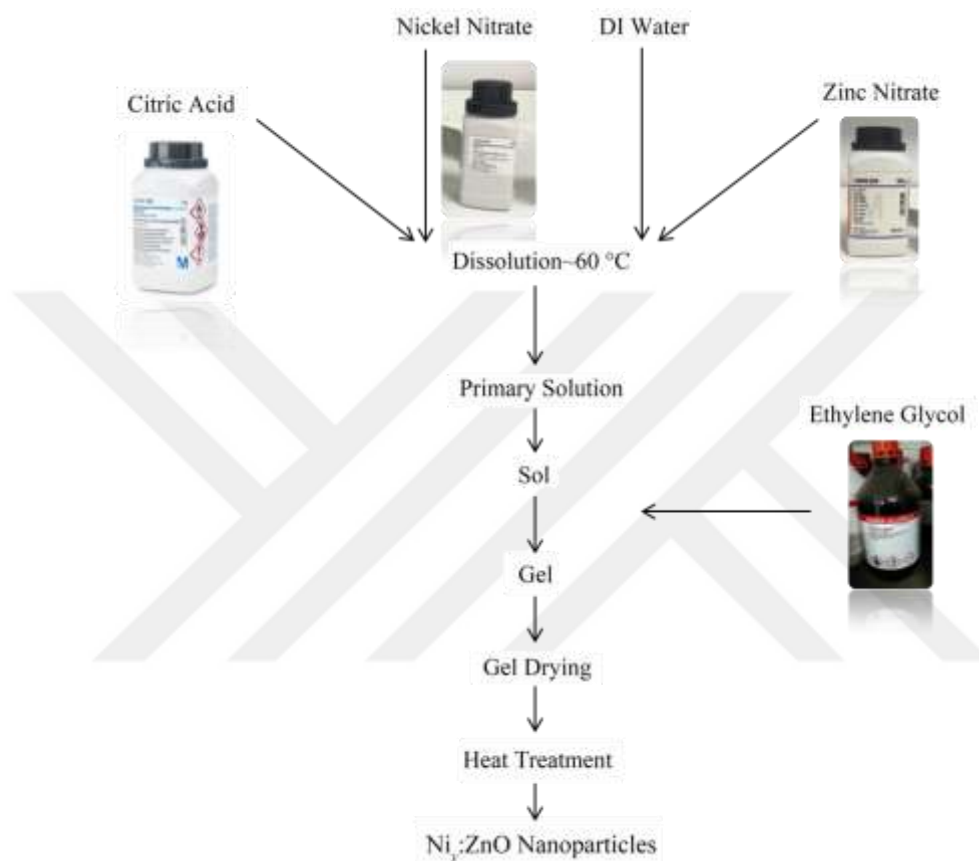


Figure 3.2. Sol-gel process of Ni_yZnO nanoparticles.

3.2.2. Synthesis of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ nanoparticles

$\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ (Nickel substituted Cobalt Ferrite) nanoparticles were synthesized by sol-gel method. In this method, 'Cobalt (II) Nitrate Hexahydrate- $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ', 'Iron (III) Nitrate Nanohydrate- $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ ', 'Nickel (II) Nitrate Hexahydrate- $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ' and 'Citric Acid' was used as starting materials. Here, Ni ratios were 0%, 25%, 50%, 75% and 100%. The starting materials dissolved in pure water in appropriate cytoichiometric proportions with a magnetic stirrer and heater at a temperature of 50-60 °C. Respectively, cobalt nitrate, iron nitrate, nickel nitrate and citric acid dissolved in pure water poured into an empty beaker and magnetic stirrer were added into the solution. The heated magnetic stirrer was set to 200-300 °C and the appropriate amount of gelling agent ethylene glycol was slowly added to the

solution. After the reaction takes place and gelation was achieved, the production process was completed. The produced $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ gels were ready for heat treatment. Figure 3.3 shows the sol-gel production scheme of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ nanoparticles.

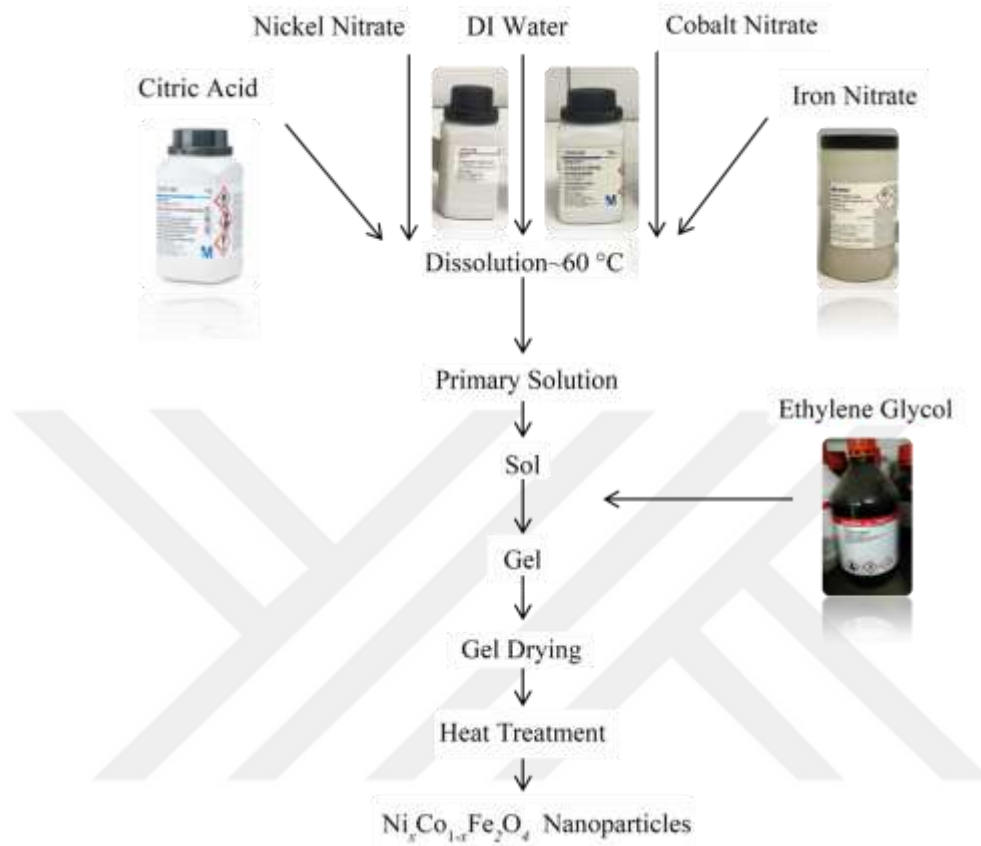


Figure 3.3. Sol-gel process of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ nanoparticles.

3.2.3. Heat treatments

In order to synthesize Ni_yZnO and $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ nanomaterials having the desired structural and magnetic properties, a controlled heat treatment process must be applied. Our preliminary studies have showed that the heat treatment process seriously affects the quality and grain size of the produced nanoparticles. For example; heat treatment of the produced samples at different temperature created different particle sizes and agglomeration. For this reason, a three-stage heat treatment method was used in this study. Figures 3.4a-c show the three-stage heat treatments that are burning, calcination and sintering, respectively. First of all, the first stage combustion heat treatment process was applied. In this process, the organic components in the material was burned. The samples heated from room temperature to 500 °C (T_1) for 20 minutes (t_1) and kept at this temperature for 40 minutes (t_2-t_1). Then it follows to cool down to room temperature (t_3-t_2) in the oven as showing in Fig. 3.4a. As a second step, a heat treatment called

calcination was applied to the burned materials. The samples heated from room temperature to 550 °C (T_2) for 40 minutes (t_4) and kept at this temperature for 260 minutes (t_5-t_4). Then, it follows to cool down to room temperature (t_6-t_5) in the oven as showing in Fig. 3.4b. After calcination process, the powder forms of materials were grinded by using agate mortar for 30 mins that it will create a fine powder. Then, the fine powders were transformed to pellet forms by using a hydrolic press. The cylindrical shape pellets having 10 mm diameter were obtained by applying 20 bar pressure. A heat treatment called sintering was applied in the third step. The samples heated from room temperature to 800 °C (T_3) for 120 minutes (t_7) and kept at this temperature for 600 minutes (t_8-t_7). Then, it follows to cool down to room temperature in the oven (t_9-t_8) as showing in Fig. 3.4c. Here, the crystallinity was obtained in the samples.



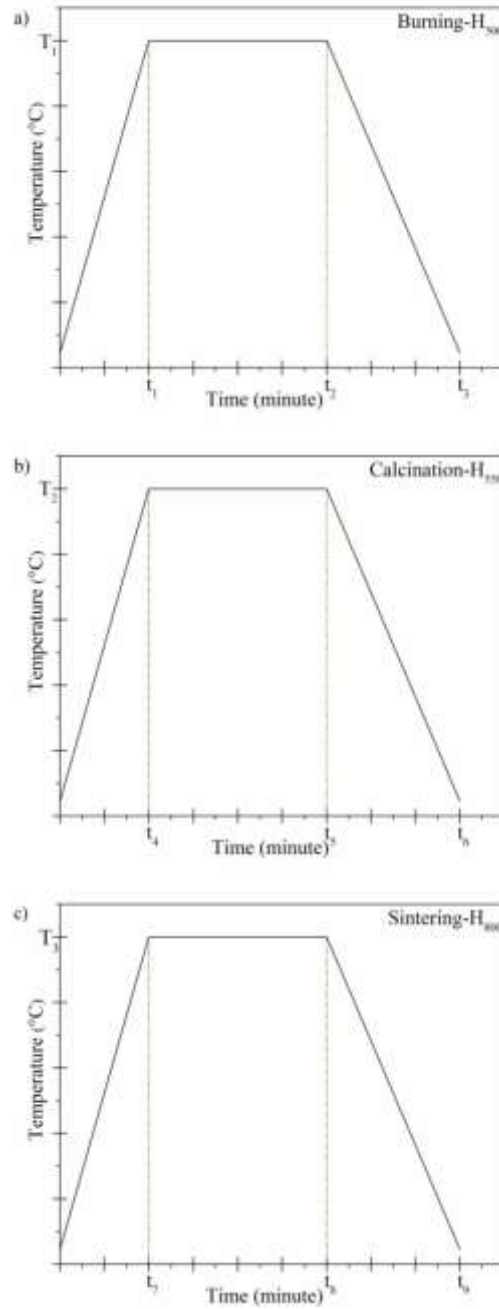


Figure 3.4. Diagram of the three stage heat treatment process: a) burning, b) calcination and c) sintering.

3.2.4. Production of radar absorbing composites (RAC)

Silicone molds with dimensions of 22.86x10.16x4 mm³ (WxLxH) was prepared to create the composite structures. The nanoparticles become homogeneous by being milled with a mechanical grinder after following the heat treatment procedures. Following the heat treatment procedures and homogenized by grinding, Ni_y:ZnO (y=0 and 0.1) and Ni_xCo_{1-x}Fe₂O₄ (x=0 to 1 with 0.25 increments) nanoparticles were mixed together with the addition of 30% by weight into the appropriate amount of epoxy/hardener. After mixing, nanomaterial particles reinforced epoxy resin was vacuumed to prevent bubbles. The epoxy/nanoparticles taken from

the vacuum was mixed again to be homogeneous. The resulting mixture was poured into silicone molds and allowed to cure for 24 hours at room temperature. Figure 3.5 shows the production stages of nanoparticle reinforced epoxy matrix composite structure.

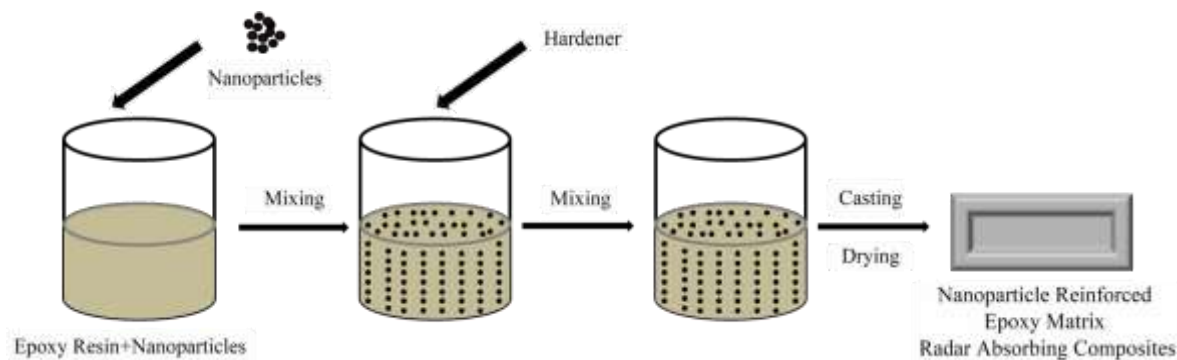


Figure 3.5. General production scheme of nanoparticle doped epoxy matrix composite.

Various composite structures were obtained by adding the produced nanomaterials into epoxy resin. In Table 3.1, the produced $\text{Ni}_y\text{:ZnO}$ and $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ nanomaterials and radar absorbing composite structures are given with their abbreviations.

Table 3.1. The abbreviations used in nanomaterials and composite structures.

Nanomaterials	Nanomaterials Codes	Composite Structures	Composite Codes
CoFe_2O_4	CNF0	$\text{ZN0}+\text{CNF0}$	RAC1
$\text{Ni}_{0.25}\text{Co}_{0.75}\text{Fe}_2\text{O}_4$	CNF1	$\text{ZN1}+\text{CNF0}$	RAC2
$\text{Ni}_{0.5}\text{Co}_{0.5}\text{Fe}_2\text{O}_4$	CNF2	$\text{ZN1}+\text{CNF1}$	RAC3
$\text{Ni}_{0.75}\text{Co}_{0.25}\text{Fe}_2\text{O}_4$	CNF3	$\text{ZN1}+\text{CNF2}$	RAC4
NiFe_2O_4	CNF4	$\text{ZN1}+\text{CNF3}$	RAC5
ZnO	ZN0	$\text{ZN1}+\text{CNF4}$	RAC6
$\text{Ni}_{0.1}\text{ZnO}$	ZN1	$\text{ZN0}+\text{CNF1}$	RAC7

	ZN0+CNF2	RAC8
	ZN0+CNF3	RAC9
	ZN0+CNF4	RAC10

Figure 3.6 represents the composite structure. The dimensions of this structure was arranged for X-band microwave experiments. After the composite structures were casting into silicon molds which were covered with 1 mm thick aluminium plate on the mold, and aluminium/nanoparticle reinforced epoxy composite structures were obtained. The purpose of aluminum plate integration is to reflect back the incoming microwaves.

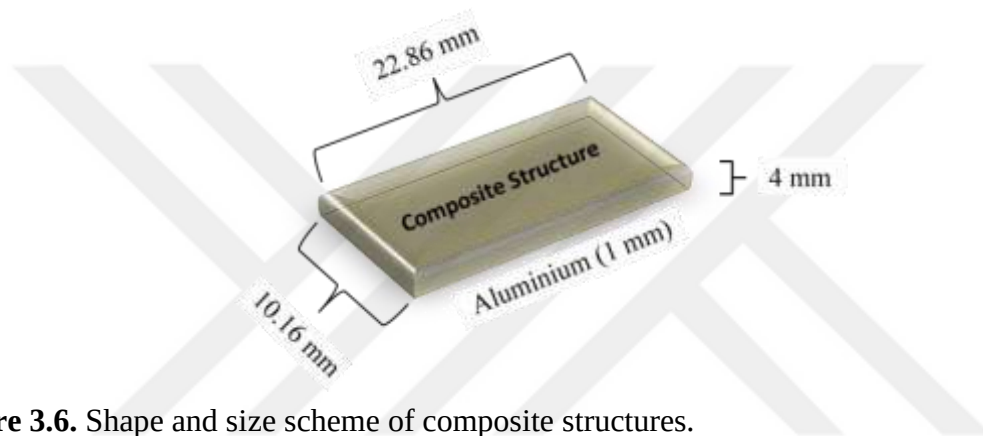


Figure 3.6. Shape and size scheme of composite structures.

Figure 3.7 shows the summary of the information about the preparation of composite structures in the thesis study by giving each step. Because each steps in the production line affects the final composite materials, it has been meticulously studied by attention to each step.



Figure 3.7. Step-by-steps from nanomaterial production to composite material production.

3.2.5. Composite sample design for mechanical tests

Physical mechanical tests applied to the radar absorber samples to be used in potential applications. Composite design was carried out in accordance with ASTM standards for the impact test and hardness test required to be tested in air, land and sea applications.

In the Charpy impact test, it was drawn with AutoCAD application in accordance with ASTM D256 standards and its volume was calculated. AutoCAD drawing shows in Figure 3.8 (All dimensions are in mm). The impact test composite sample was prepared by casting an appropriate amount of nanoparticles reinforced epoxy resin into the mold made of wood.

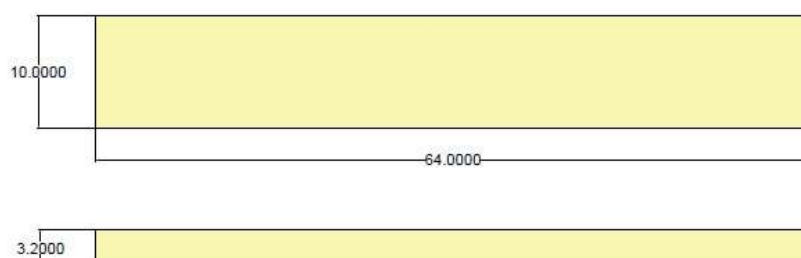


Figure 3.8. Impact test specimen dimensions in accordance with ASTM D256 standards.

In the hardness test, it was drawn with AutoCAD drawing program in accordance with ASTM D785 standards and its volume was calculated. Sample dimensions accordance with ASTM D785 standards given in Figure 3.9 (dimensions in mm). The hardness test composite sample was prepared by casting an appropriate amount of nanoparticles reinforced epoxy resin into the mold made of wood.

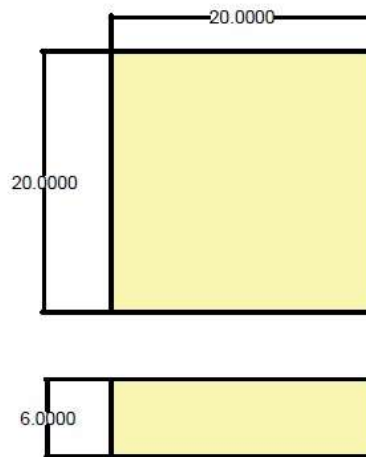


Figure 3.9. Hardness test specimen dimensions in accordance with ASTM D785 standards.

The images of the molds of impact and hardness test samples prepared in accordance with ASTM D256 and ASTM D785 standards are given in Figure 3.10. There are many reasons for wooden molds are preferred such as easy formability, affordable cost, precise measurements and not damaging the material.

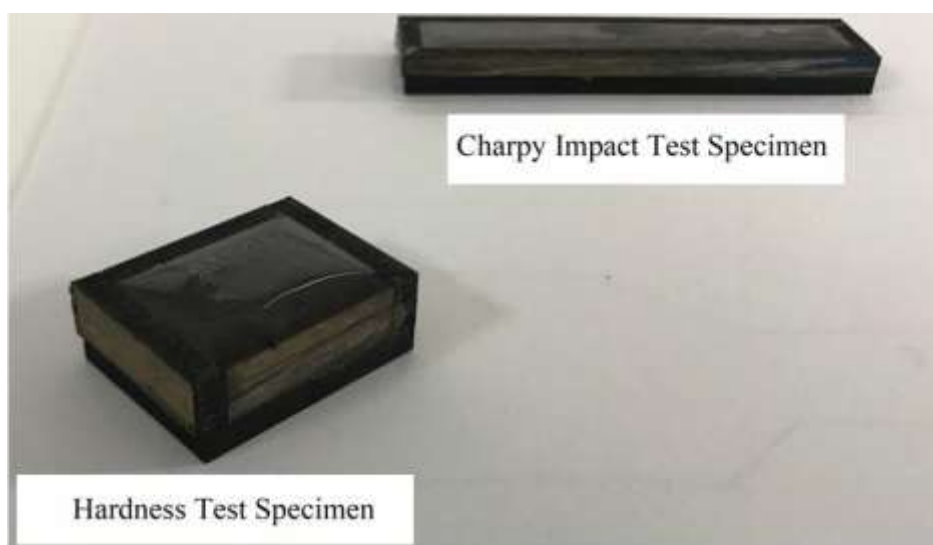


Figure 3.10. Mold samples prepared from wood and epoxy casting for mechanical tests.

3.3. Characterization techniques

Some physical (structural, morphological, elementary, mechanic, magnetic and electromagnetic) properties of nanoparticles and composite structures were studied by using various characterization techniques. In the following section, we will describe each characterization techniques.

3.3.1. Structural analysis

X-ray diffraction (XRD), scanning electron microscopy (SEM) and energy dispersive x-ray spectrometry (EDS) measurements were performed to analyze the structural properties of the Ni_yZnO and $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ nanoparticles.

3.3.1.1. X-ray diffraction (XRD) method

The XRD characterization method is based on the diffraction of the x-rays sent on the sample. It allows the determination of particle size, crystal size, lattice planes and crystal phase of materials. German Physicist Max von Laue found that when the atoms in the sample structure were in crystalline form, the X-ray sent to the structure was refracted. Laue conducted studies on copper-sulphate crystal to record these diffractions. Laue wanted to prove that the atoms were arranged in a periodic structure in the crystals with x-rays which sent on copper sulphate. Laue won the Nobel Prize in Physics in 1914 as a result of this experiment. This work of Laue attracted attention by W. L. Bragg and was expressed mathematically under the name of "Bragg's Law". The mentioned mathematical expression can be expressed as follows.

$$n\lambda = 2d\sin\theta \quad (22)$$

In this equation, n represents an integer. λ is the wavelength of the incident beam, θ is the angle that it makes with the crystal plane and d is the distance between the two planes in the crystalline structure. The Bragg rule satisfies this condition in the case of $\lambda \leq 2d$ (λ is equal and smaller than $2d$). Bragg reflection occurs when this condition is suitable.

Figure 3.11 shows a schematic illustration of an x-ray diffraction from a crystallized material. The path difference of the scattered rays between two consecutive planes must be exactly multiple layers of the wavelength to occur a constructive interference. The reflected rays in this way are in the same phase and reinforce each other to form diffraction rays.

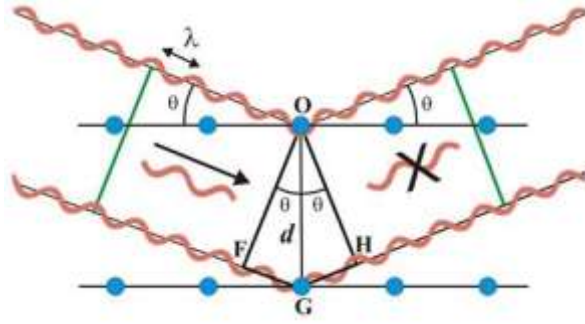


Figure 3.11. X-ray diffraction and Bragg's law on crystal planes ("Bragg Law," 2020).

In the x-ray diffraction method, the crystal samples to be measured are formed into a homogeneously distributed very fine powder and placed in a single wavelength x-ray. Maximum reflection can be achieved when the Bragg angle is correct and the scattered rays reach the detector. In the x-ray diffraction method, a graph of the intensity of diffracted rays are given as a function of the (2θ) reflection angle. In this study, x-ray diffraction measurements of the samples were made with a Cu-K α (1.5405 Å) x-ray source and PANalytical EMPYREAN x-ray diffraction analysis system at Çukurova University Central Research Laboratory (ÇÜMERLAB) (see Figure 3.12). Measurements were implemented between 20° and 80° in 0.002° steps at room temperature.



Figure 3.12. X-ray diffraction system of PANalytical EMPYREAN.

3.3.1.2. Scanning electron microscopy (SEM) and energy-dispersive x-ray spectrum (EDS) methods

SEM is used to characterize the surface and intersections by analyzing the morphology of the samples. Figure 3.13 shows the schematic working principle of the scanning electron microscope. The electron gun at the top of the SEM generates the electron beam. The generated electron beam passes through lenses. Lenses are used to collect the electron beam and focus it on the sample. Focusing on the sample, the impinging electron beam interacts with the atoms in the sample and scatters back. A high resolution image is created by collecting scattered signals from different regions with the detectors in the SEM system.

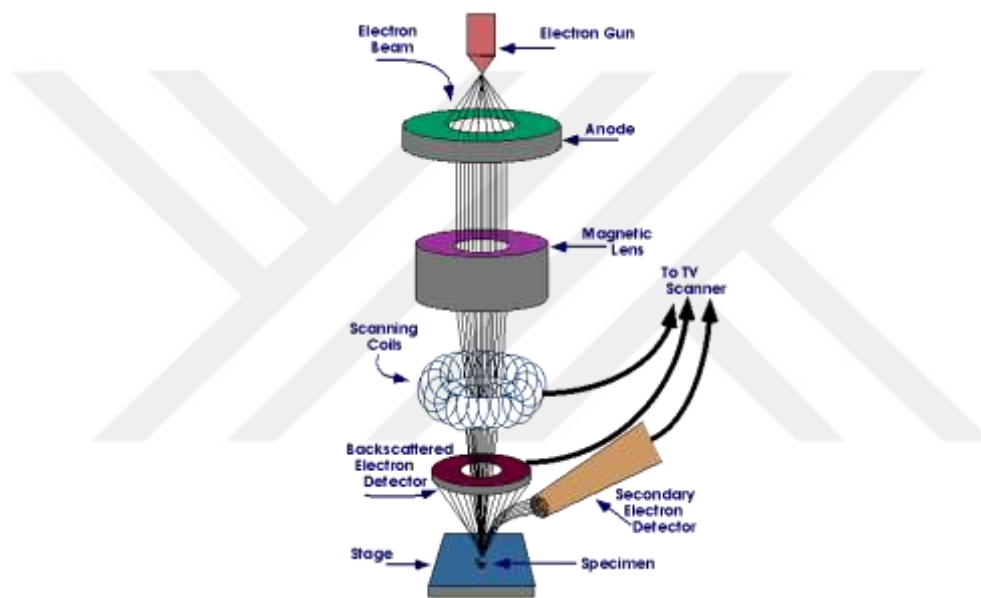


Figure 3.13. Scanning electron microscope working principle scheme (SEM, 2020).

The EDS analysis is used to determine the elements and their quantities present in material. Similar to the SEM method, EDS is based on the scattering of high-energy electrons by hitting the sample surface. The scattered waves have characteristic features depending on the energy and wavelengths. Accordingly, the analysis and quantities of the elements in the sample are determined. SEM images and EDS analysis of nanoparticles produced in this study were obtained by EDS integrated FEI Quanta FEG 650 model scanning electron microscope at Çukurova University Central Research Laboratory (see Figure 3.14).



Figure 3.14. FEI Quanta FEG-650 type scanning electron microscope with EDS integrated.

3.3.2. Magnetic analysis

Physical Property Measurement System (PPMS, Quantum Design-DynaCool) is the measurement system used to determine the electrical, magnetic and thermal properties of materials. There are different modules in this system. One of these modules is the Vibrating Sample Magnetometer (VSM) used to determine the magnetic properties of the samples.

In the thesis study, all of the magnetic analysis of nanoparticle samples were made by the Quantum Design brand DynaCool model PPMS system in ÇÜMERLAB (see Figure 3.15). The operating temperature of the PPMS system is in the range of 1.85 K-400 K and analyzes are performed under ± 9 T magnetic field. In temperature dependence magnetization experiments, zero field cooling (*ZFC*) and field cooling (*FC*) processes were applied for $\text{Ni}_y\text{:ZnO}$ samples. Magnetic hysteresis curves for $\text{Ni}_y\text{:ZnO}$ samples were measured at 5K and 300K temperatures and for $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ samples at 300K.



Figure 3.15. Quantum Design Dyna-Cool model Physical Property Measurement Sytem (PPMS).

3.3.3. Electromagnetic analysis

Network analyzer is an important system for the analysis, control and monitoring of components and devices to be used in microwave (*RF*) systems. Network analyzer provides to measure of network scattering parameters, reflection measurements and analysis of S parameters of samples.

The electromagnetic properties of the RAC samples specified in Table 3.1 were studied in the frequency range of 8 GHz-12 GHz, called as X band. Composite samples for reflection loss measurements are manufactured in standard waveguide size of X-band as 22.86 mm (0.9 inch) long and 10.16 mm (0.4 inch) wide. In order to provide full reflection in the analysis part, aluminum sheet having 1 mm thickness is integrated back to the samples in composite production. Thus, the reflection of the electromagnetic waves passing through the sample is provided and it is provided to pass through the composite sample again. The purpose here is to calculate the energy loss at the entrance and exit of the wave and the absorption capacity of the structure. In the thesis study, all of the electromagnetic analysis of composite samples were made by the KEYSIGHT PNA-L Network Analyzer system in İskenderun Technical University (See Figure 3.16). Reflection loss graphs in the range of 8 GHz and 12 GHz frequencies were obtained in dB.

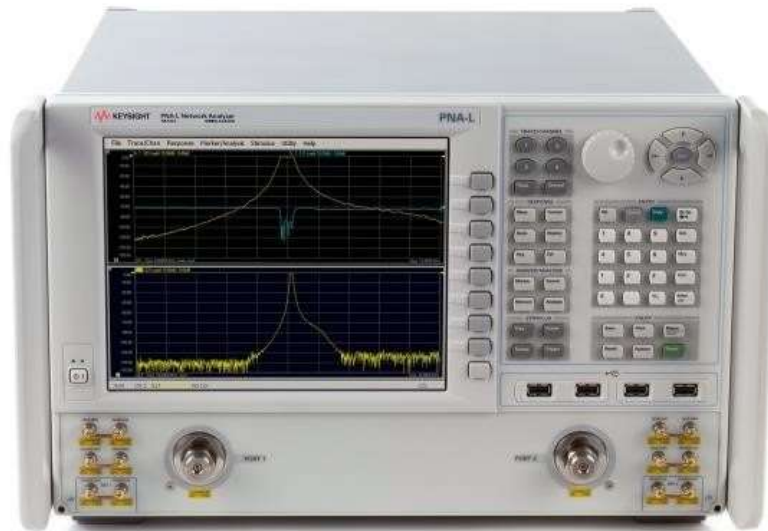


Figure 3.16. PNA-L type KEYSIGHT network analyzer system.

3.3.4. Mechanical analysis

The nanoparticles reinforced epoxy matrix radar absorber composite structures were further analyzed to determine their physical properties, structural integrity and mechanical strength. Composite samples have been designed, produced and analyzed in accordance with certain standard procedures (ASTM). Charpy impact test and hardness test analyzes were performed on composite specimens.

3.3.4.1. Charpy impact test

Impact test is used to determine the behavior of the material exposed to sudden load and to determine its resistance against dynamic stresses. Impact resistance affects product life and reliability in potential applications of composite specimens. The purpose of the Charpy impact test is to learn the fracture stages of the materials and to determine their impact resistance.

In the charpy impact test, all the energies required to initiate the crack, ensure its progress and create an elastic and plastic deformation zone represent the total energy in the charpy test. Specimen was prepared in 10 mm width, 64 mm length and 3.2 mm thickness in accordance with the ASTM D256 standard. The impact energy was recorded by the MITECH XJJ-50S Digital Charpy Impact Test Analyzer given in the Fig. 3.17.



Figure 3.17. MITECH XJJ-50S digital charpy impact tester.

3.3.4.2. Hardness test

Hardness is the ability of the material to resist scratching and plastic deformation. The ability to resist plastic deformation is related to yield strength. The hardness test is measured by the resistance of the material by dipping a standard indenter into the surface of the material. The tips submerged in the surface of the material vary according to the test sets.

The sample to be used in the hardness test was prepared in accordance with ASTM D785 standards, with a corner of 20 mm in length and 6 mm in thickness. Hardness test was applied in ASTM D785 standards. LOYKA Digital Shore D Shoremeter hardness test analyzer was used in the hardness measurement test (Fig. 3.18).



Figure 3.18. LOYKA Digital Shore D Durometer hardness test analyzer.

4. RESULTS AND DISCUSSIONS

In this thesis, $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ ($0.0 \leq x \leq 1.0$ with 0.25 increments) and $\text{Ni}_y\text{:ZnO}$ ($y=0$ and 0.1) nanoparticles were produced by using sol-gel method and three stages heat treatment process were applied to the nanoparticles as explained in Chapter 3. The produced nanoparticles were reinforced into epoxy resin as indicated in Table 3.1 and various radar absorbing composite structures were obtained. Structural and magnetic characterizations of synthesized nanoparticles are discussed in Section 4.1 and Section 4.2, respectively. The electromagnetic (reflection losses) properties of nanoparticles reinforced radar absorbing composite structures are discussed in Section 4.3. Finally, the mechanical properties of composite structures are studied in Section 4.4.

4.1. Structural properties of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ ($0.0 \leq x \leq 1.0$) and $\text{Ni}_y\text{:ZnO}$ ($y=0$ and 0.1) nanoparticles

4.1.1. X-ray diffraction (XRD) analysis of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ ($0.0 \leq x \leq 1.0$) and $\text{Ni}_y\text{:ZnO}$ ($y=0$ and 0.1) nanoparticles

Synthesized nanoparticles are characterized by powder X-ray diffraction (XRD) measurement method for structural determination. Before XRD analysis, the samples are grinded to get their powder forms. XRD measurements were performed by sweeping the diffraction angles between 20° and 80° . After acquiring XRD datas various structural parameters such as crystallite sizes, phase determination, lattice parameters were obtained using X' Pert HighScore Plus software.

Figures 4.1a-e show the XRD patterns of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ nanoparticles for $x=0.00$, $x=0.25$, $x=0.50$, $x=0.75$ and $x=1.00$, respectively. The X-Pert High-Score Plus software has confirmed that the nanoparticles have a single cubic crystal structure with the $Fd 3m$ space group. The samples appear to exhibit characteristic $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ diffraction peaks. The intensities of major diffraction peaks of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ samples increase with increasing nickel content as seen in Figs.1a-e. In addition to characteristic cubic spinel crystal peaks, the $\alpha\text{-Fe}_2\text{O}_3$ impurity phase as indicated by the star symbol belonging to the synthesized nanoparticles was found in all samples. The formation of impurity peaks depend on experimental parameters such as the composition of the precursors, the pH of the solution, and the rate of heating/cooling during synthesis (Raja et al., 2015).

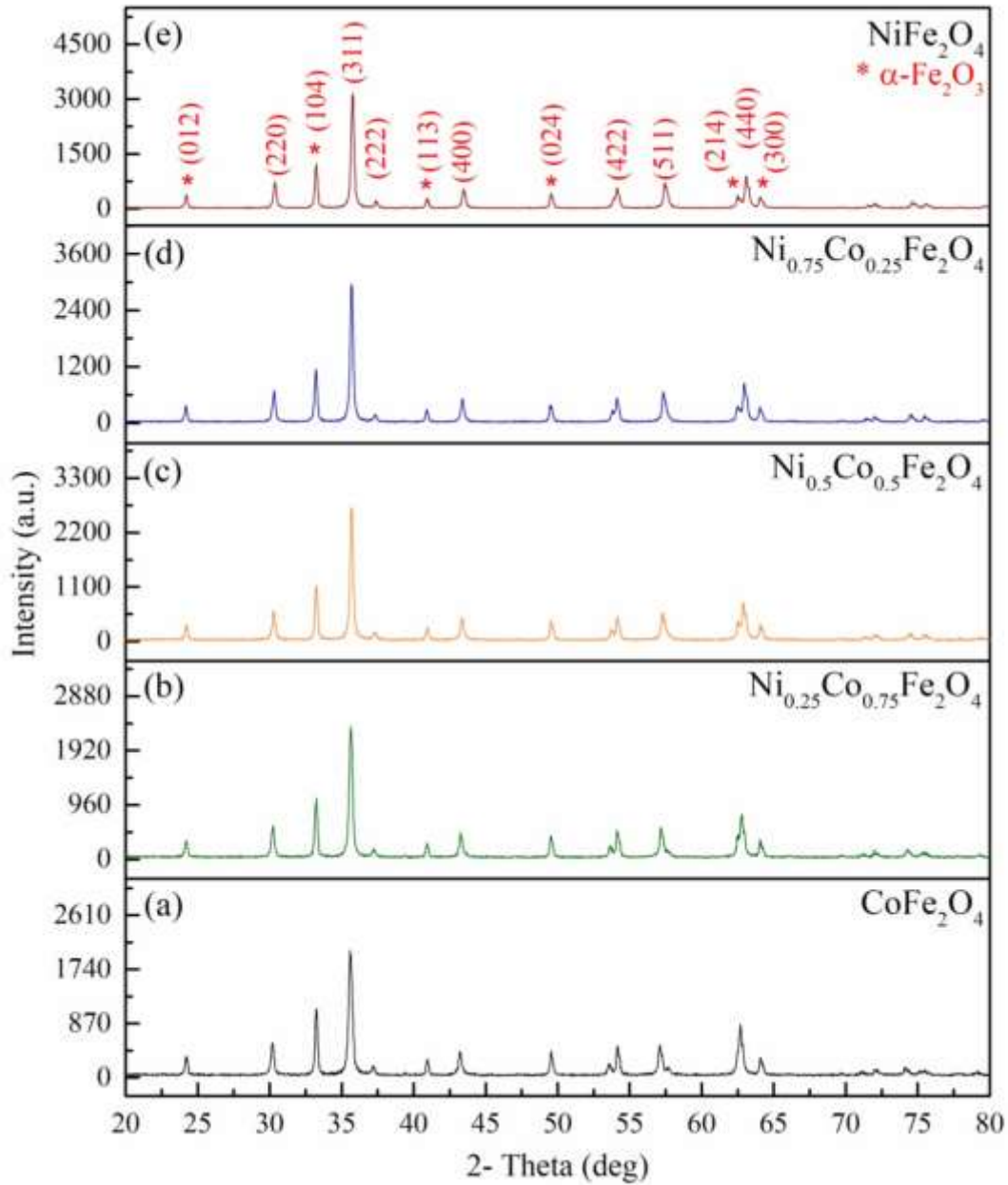


Figure 4.1. X-ray diffraction patterns of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ nanoparticles for a) $x=0.00$ b) $x=0.25$ c) $x=0.50$ d) $x=0.75$ and e) $x=1.00$.

The lattice parameters of the samples were found to be 8.361, 8.355, 8.343, 8.342 and 8.323 for $x=0.00$, 0.25, 0.50, 0.75 and 1.00, respectively that it decreases with increasing Ni^{+2} content in $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ as shown in Figure 4.2. Since the ionic radius of Ni^{+2} (0.63 Å) is smaller than Co^{+2} (0.78 Å), the lattice parameter decreases when increasing of Ni^{+2} content in the structure (Kumar et al., 2015). A slight shift towards the lower angle sides is observed when Ni amount is increased in the Co-ferrite structure (El-Sayed, 2002). This situation is due to the small ionic radius difference between Ni^{+2} and Co^{+2} .

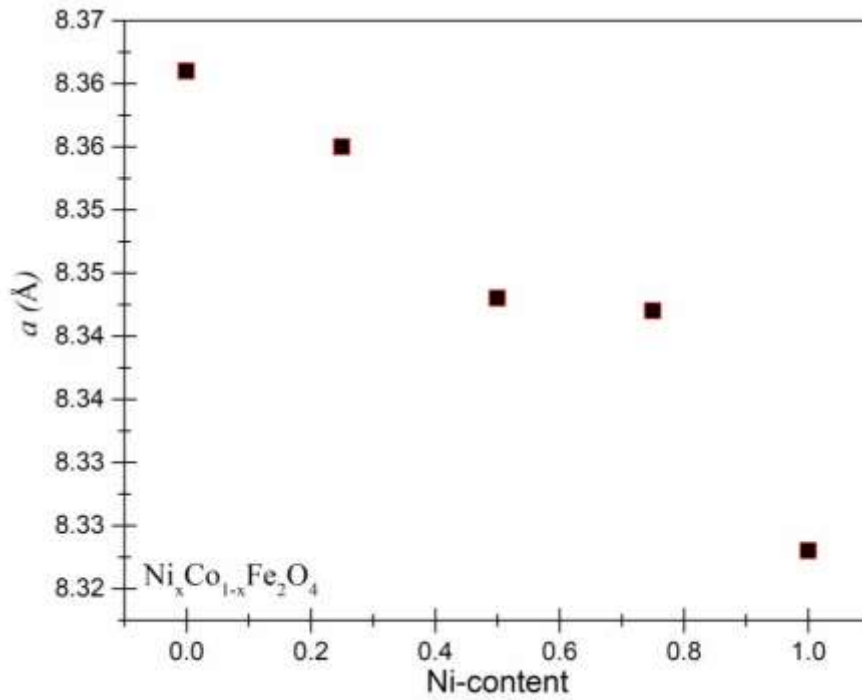


Figure 4.2. Lattice parameter (a) as function of Ni-content in $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ samples.

Further structural properties were investigated by calculating the crystallite size of the samples using the Debye-Scherrer equation (Cullity, 1956).

$$D = \frac{\lambda \kappa}{\beta \sin \theta} \quad (23)$$

where D is the crystallite size, κ is the shape factor (0.9), λ is the x-ray wavelength ($\text{CuK}\alpha=1.5406 \text{ \AA}$), β is the fullwidth at half maximum of the diffraction peaks and θ is the Bragg angle. The calculated crystallite size of the $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ samples are found as 30.68 nm, 36.61 nm, 44.92 nm, 44.93 nm and 42.73 nm for $x=0.00$, 0.25, 0.50, 0.75 and 1.00, respectively. Crystallite size of samples increase with increasing nickel content up to $x=0.75$ and then decreased with further nickel content. This situation is consistent with the literature due to the ionic radius of Ni^{2+} and Co^{2+} ions (He, 2014).

Table 4.1. Structural properties of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ samples.

Sample Code	Lattice Parameter a (Å)	Unit Cell Volume V (Å ³)	d -spacing value (Å)	Crystallite Size D (nm)
CNF0	8.3605	586.77	2.5208	30.68
CNF1	8.3550	583.24	2.5191	36.61
CNF2	8.3429	580.69	2.5155	44.92
CNF3	8.3424	580.59	2.5153	44.93
CNF4	8.3279	577.57	2.5110	42.73

Figures 4.3a-b show the XRD patterns of Ni_y:ZnO nanoparticles for $y=0$ and $y=0.1$, respectively. Characteristic hexagonal crystal diffraction peaks are observed in the XRD spectrum. The X-Pert High Score Plus software has confirmed that the ZnO diffraction peaks have a hexagonal crystal structure with $P6_3mc$ space group. A second crystal as formed NiO indicated by a hashtag symbol is observed in Fig. 4.3b NiO diffraction peaks have cubic crystal structure with $Fm-3m$ space group.

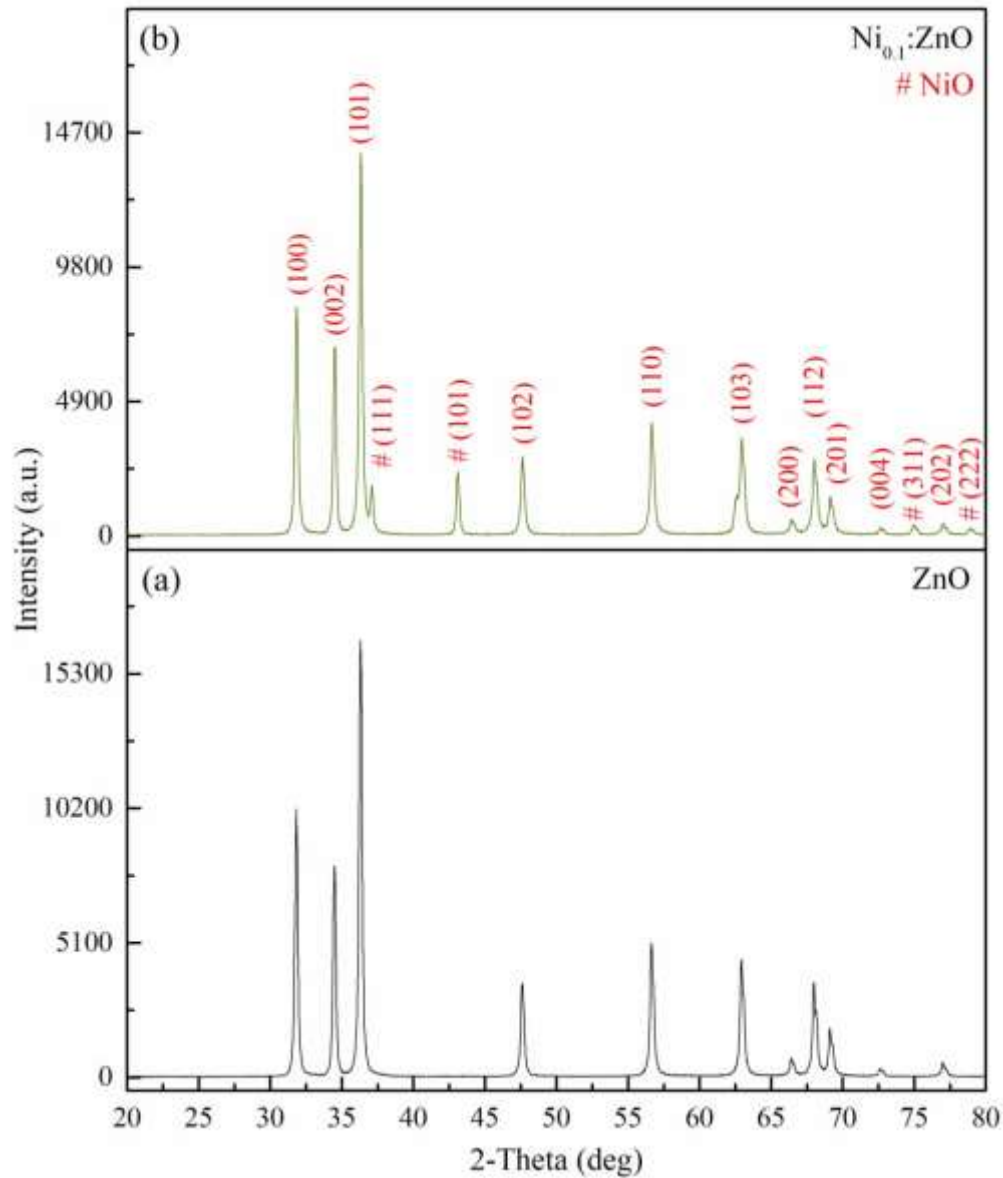


Figure 4.3. X-ray diffraction patterns of Ni_yZnO nanoparticles for a) $y=0.00$ and b) $y=0.10$.

The crystallite sizes of ZN0 and ZN1 samples calculated by using Eq.23 are found almost same. This confirms that the Ni ions do not affect the lattice of main structure. The lattice parameters such a and c of the ZN0 (ZN1) sample were found as 3.2473 Å (3.2457 Å) and 5.2034 Å (5.2019 Å), respectively. The calculated crystallite size from the XRD results of the ZN0 and ZN1 samples are found as 40.42 nm and 40.22 nm, respectively. A slight decrease in lattice parameter and crystallite size of samples might be related to adding Ni^{2+} in to the main ZnO structure. Although Ni^{2+} ions are added in to the ZnO structure, the decrease in crystal size and lattice parameter indicates a possibility of substitution the Ni^{2+} ion (0.690 Å) with Zn^{2+} ion (0.740 Å). The reason for this could be related with smaller ionic radius of Ni^{2+} ion than Zn^{2+} ion.

Table 4.2. Structural properties of Ni_yZnO samples.

Sample Code	Lattice Parameter		Unit Cell value (Å)	Unit Cell Size	d -spacing a (Å)	Crystallite	
	c/a	Volume D (nm)				c (Å)	V (Å ³)
ZN0	3.2473	5.2034	1.602	53.246	2.4740	40.42	
ZN1	3.2457	5.2019	1.603	53.171	2.4729	40.22	

4.1.2. Scanning electron microscope (SEM) and energy-dispersive x-ray spectrum (EDS) analysis of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ ($0.0 \leq x \leq 1.0$) and Ni_yZnO ($y=0$ and 0.1) nanoparticles

Microstructural properties of the produced samples were examined by scanning electron microscope (SEM) images at different magnifications. In all samples, the grain size distribution plottings were determined by imageJ software based on 100 randomly selected grains from the SEM image.

SEM images of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$, $0.0 \leq x \leq 1.0$ with 0.25 increment, nanoparticles taken at 20 kX and 50 kX magnifications are given in Figs. 4.4 (a-b), 4.5 (a-b), 4.6 (a-b), 4.7 (a-b) and 4.8 (a-b), respectively. In all SEM figures, the nanoparticles appear to be agglomerated by heat treatment and to be formed as nearly spherical shape. Since agglomerations in the structures contribute to the electromagnetic absorption by decreasing the wave energy, this situation might enhance the absorption of electromagnetic waves. The average particle size of the nanoparticles were found as 0.161, 0.159, 0.165, 0.214 and 0.155 μm for CNF0, CNF1, CNF2, CNF3 and CNF4 samples, respectively (see Figs. 4.4 (c), 4.5 (c), 4.6 (c), 4.7 (c). and 4.8 (c)). The average particle size of samples are found larger than the crystal sizes obtained in XRD analysis.

Energy dispersive x-ray spectrums (EDS) of the CNF0, CNF1, CNF2, CNF3 and CNF4 samples are shown in Figs. 4.4 (d), 4.5 (d), 4.6 (d), 4.7 (d) and 4.8 (d), respectively. As can be seen from the EDS spectrum, there were no impurities and the compounds were formed accurate. Inset of Figs. 4.4 (d), 4.5 (d), 4.6 (d), 4.7 (d) and 4.8 (d) indicates the weight and atomic percentages of the elements in the compound. Table 4.3 showed that atomic percentages of each element based on desired and EDS results of the all samples.

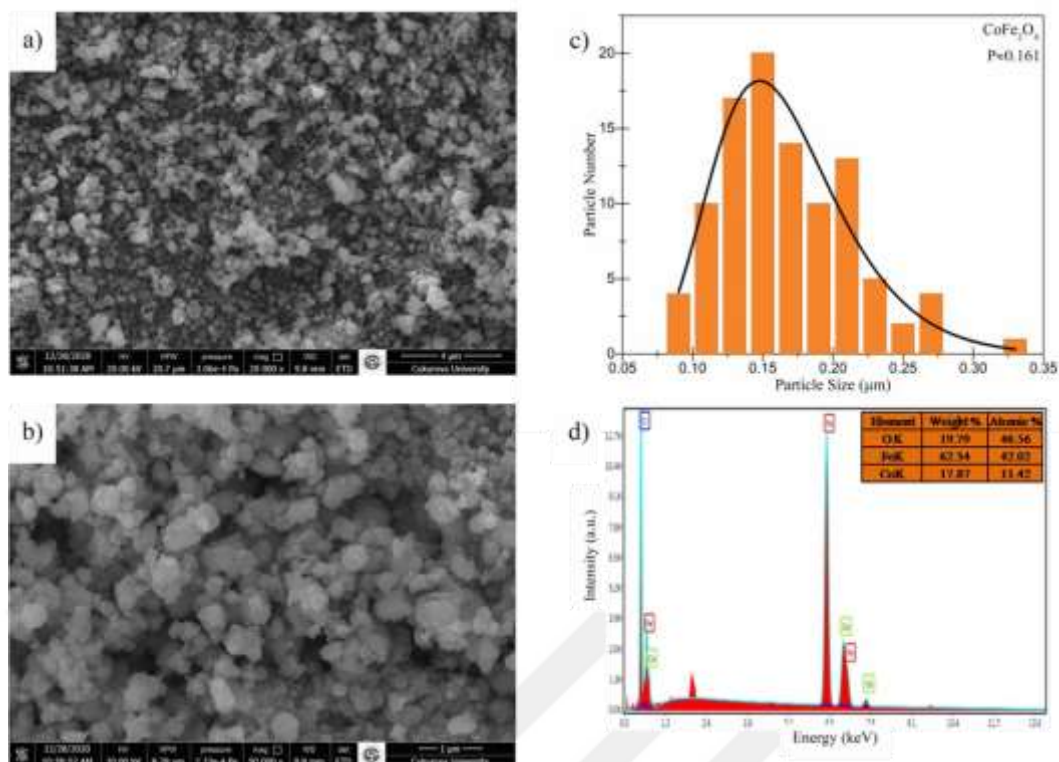


Figure 4.4. CNF0 sample; a-b) SEM images, c) particle size distribution plot and d) EDS spectrum (inset of d, atomic and weight percentage of elements).

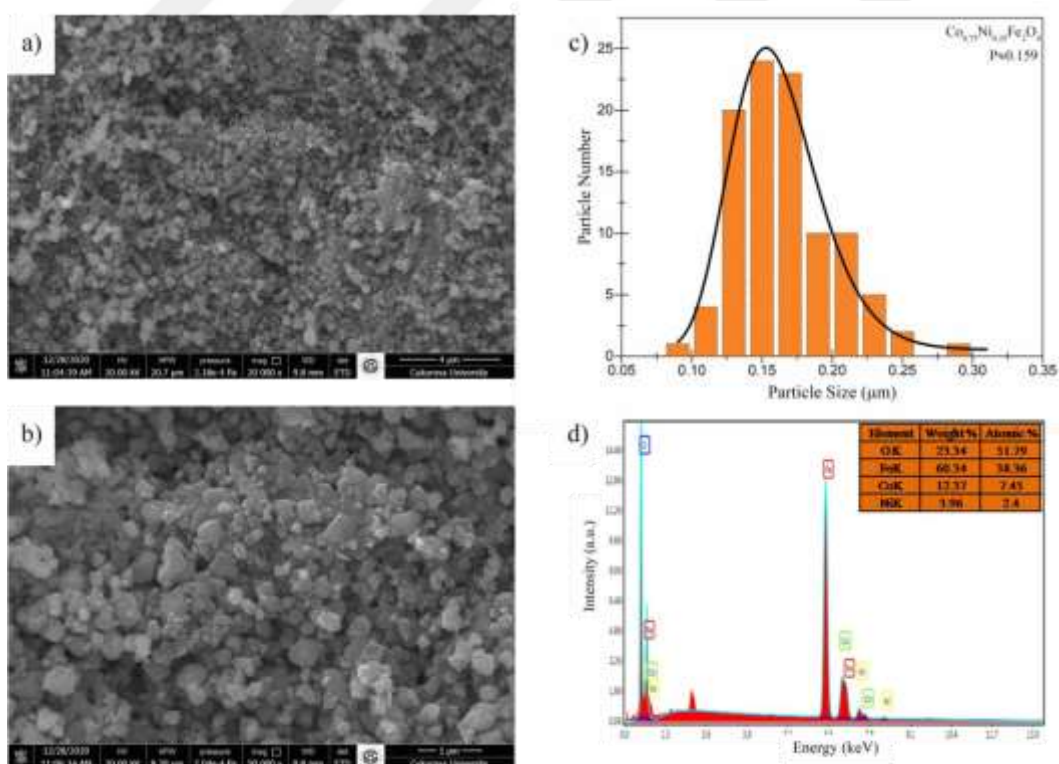


Figure 4.5. CNF1 sample; a-b) SEM images, c) particle size distribution plot and d) EDS spectrum (inset of d, atomic and weight percentage of elements).

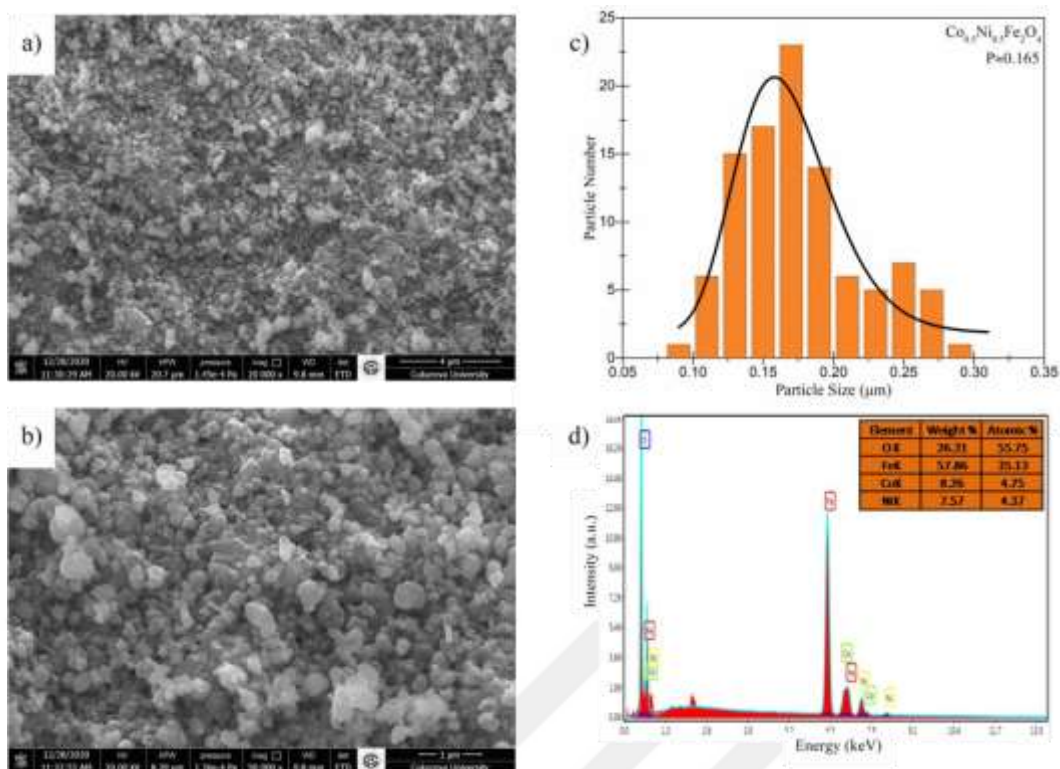


Figure 4.6. CNF2 sample; a-b) SEM images, c) particle size distribution plot and d) EDS spectrum (inset of d, atomic and weight percentage of elements).

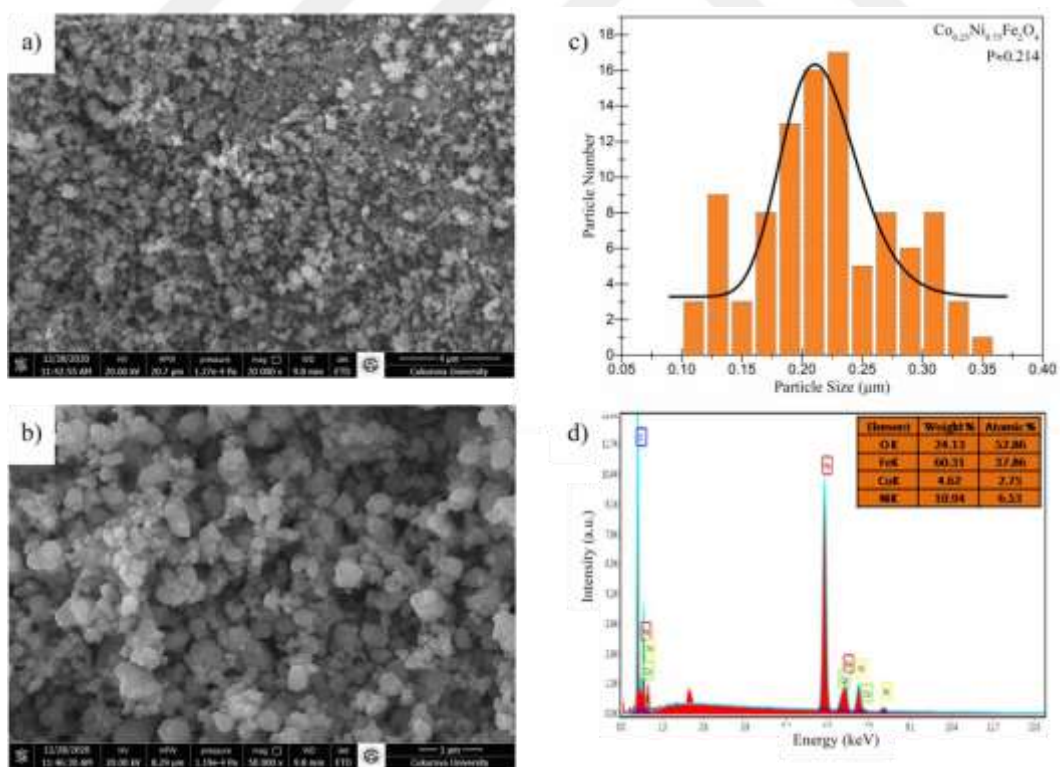


Figure 4.7. CNF3 sample; a-b) SEM images, c) particle size distribution plot and d) EDS spectrum (inset of d, atomic and weight percentage of elements).

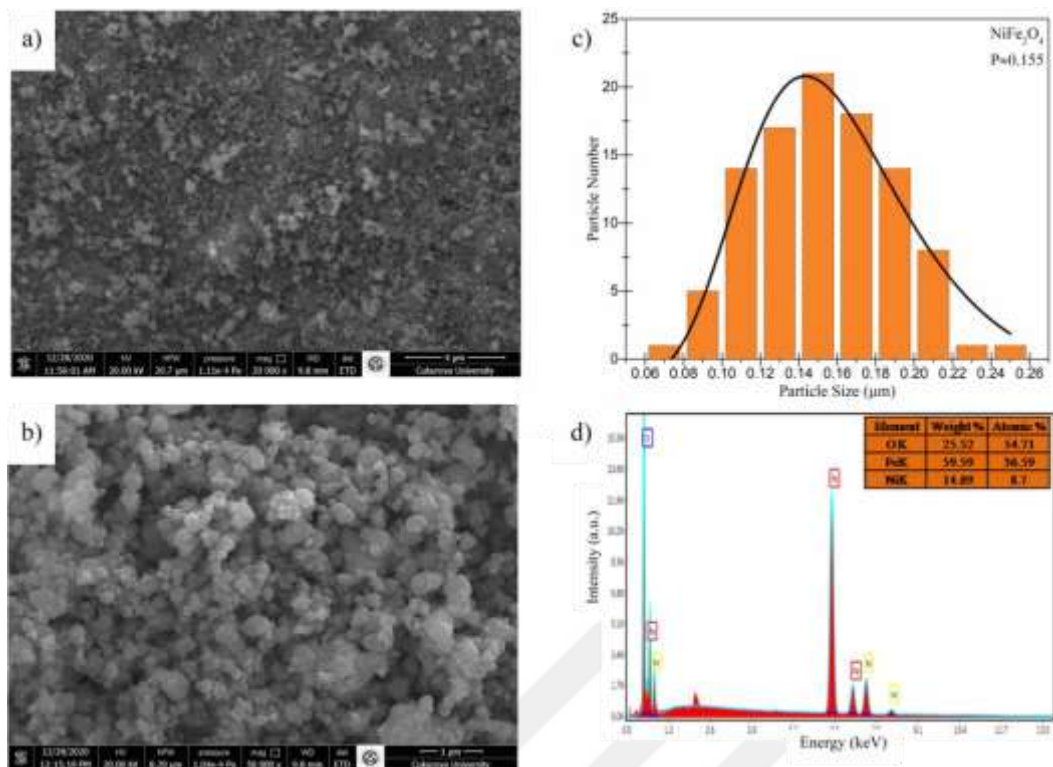


Figure 4.8. CNF4 sample; a-b) SEM images, c) particle size distribution plot and d) EDS spectrum (inset of d, atomic and weight percentage of elements).

Figure 4.9 shows the average particle size depending on Ni content. In CNF0, CNF1, CNF3 and CNF4 samples, the average particle sizes were distributed in the range of 0.155-0.165 μm , while the average particle size for CNF3 was around 0.214 μm . The reason of the average grain size in the CNF3 sample was different from the other samples may be related to the experimental conditions.

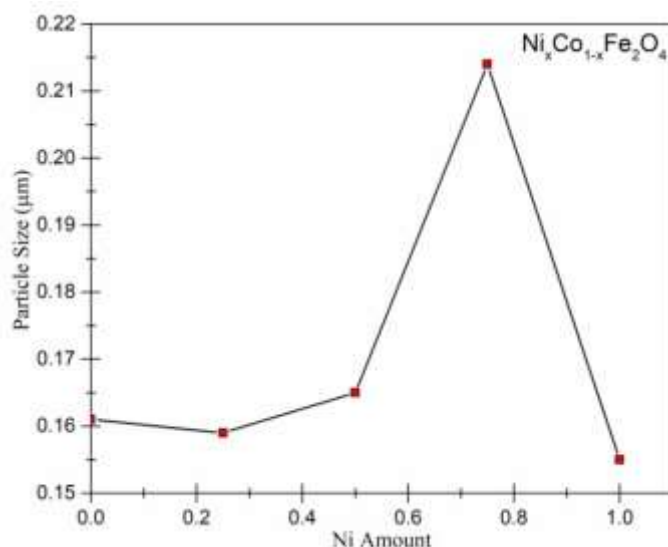


Figure 4.9. Average particle size as a function of Ni amount in $Ni_xCo_{1-x}Fe_2O_4$ structure.

Table 4.3. The desired and EDS results of atomic percentage of each element in $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ samples.

Sample Code	Desired				Measured (EDS)			
	Co	Ni	Fe	O	Co	Ni	Fe	O
CNF0	1.00	0.00	2.00	4.00	0.80	0.00	2.94	3.26
CNF1	0.75	0.25	2.00	4.00	0.53	0.17	2.68	3.62
CNF2	0.50	0.50	2.00	4.00	0.33	0.31	2.46	3.90
CNF3	0.25	0.75	2.00	4.00	0.19	0.46	2.65	3.70
CNF4	0.00	1.00	2.00	4.00	0.00	0.61	2.56	3.83

SEM images of Ni_yZnO ($y=0$ and 0.1) samples were taken at 20 kX and 50 kX magnifications for more accurate analysis of the particles as seen in Figs. 4.10 (a-b) and 4.11 (a-b), respectively. The grains were clearly visible in both micrographs. Nanoparticles were observed in the form of a sphere and agglomerated region by region. The average particle size of the ZN0 and ZN1 samples were found around 0.234 and $0.177 \mu\text{m}$, respectively (see Figs. 4.10 (c) and 4.11 (c)). Particle size decreased from 0.234 to $0.177 \mu\text{m}$ with the increase in the amount of Ni. The average particle size of the ZN0 and ZN1 samples are larger than the crystal sizes obtained in XRD analysis.

Energy dispersive x-ray spectrums (EDS) of the ZN0 and ZN1 samples are shown in Figs. 4.10 (d) and 4.11 (d), respectively. As can be seen from the EDS spectrum, there were no impurities and the compounds were formed accurate. Insets of Figs. 4.10 (d) and 4.11 (d) indicate the weight and atomic percentages of the elements in the compound. Table 4.4 shows the atomic percentages of each element based on desired and EDS results of the all samples.

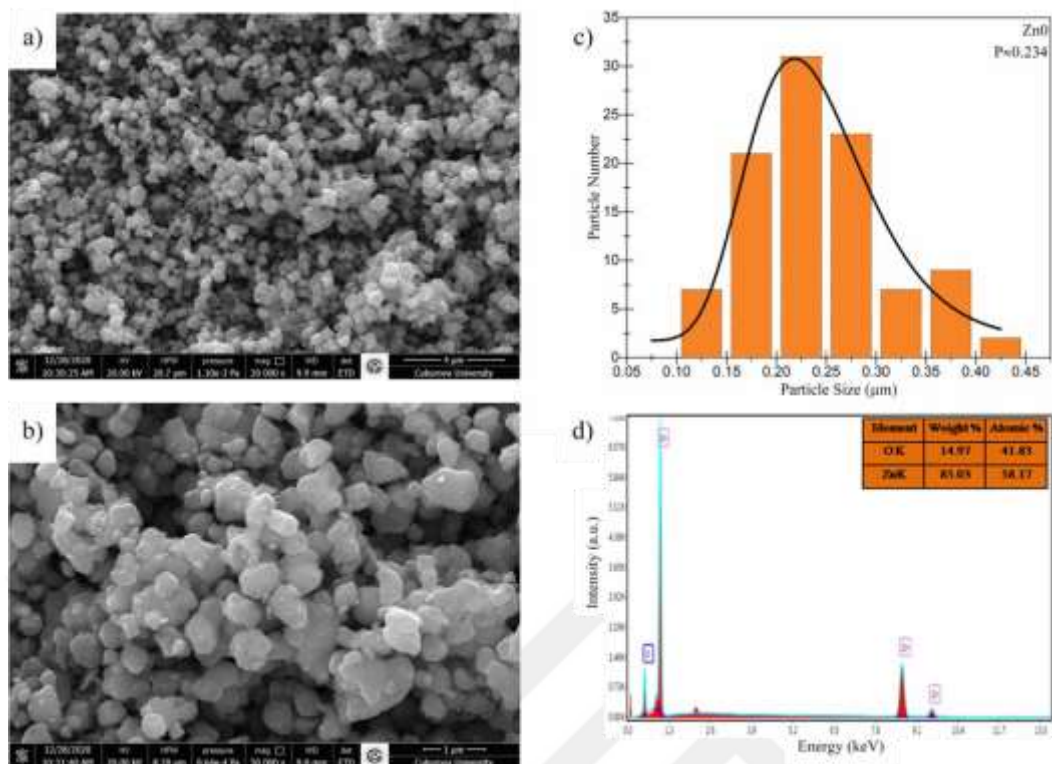


Figure 4.10. ZnO sample; a-b) SEM images, c) particle size distribution plot and d) EDS spectrum (inset of d, atomic and weight percentage of elements).

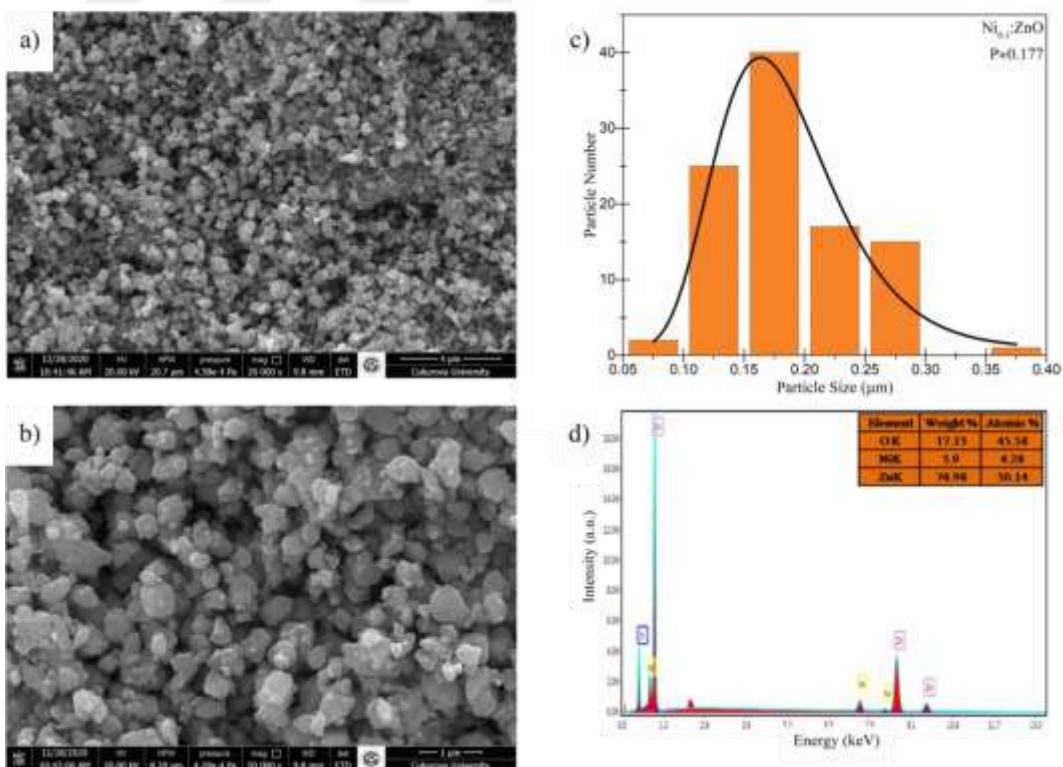


Figure 4.11. Zn1 sample; a-b) SEM images, c) particle size distribution plot and d) EDS spectrum (inset of d, atomic and weight percentage of elements).

Table 4.4. The desired and EDS results of atomic percentage of each element in Ni_yZnO samples.

Sample Code	Desired			Measured (EDS)		
	Ni	Zn	O	Ni	Zn	O
Zn0	0.00	1.00	1.00	0.00	0.84	1.16
Zn1	0.10	1.00	1.00	0.09	1.00	0.91

4.2. Magnetic properties of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ ($0.0 \leq x \leq 1.0$) and Ni_yZnO ($y=0$ and 0.1) nanoparticles

4.2.1. Temperature dependence of magnetic properties of Ni_yZnO nanoparticles The temperature dependent magnetization (M - T) values of the produced Ni_yZnO samples were measured by sweeping temperature in the range of 5-300 K under magnetic field of 250 Oe. The M - T datas were collected in both zero-field-cooled (ZFC) and field-cooled (FC) conditions. In ZFC measurements, the samples were cooled from room temperature to 5K under zero field, and then moment datas were collected while the samples were warmed under a 250 Oe magnetic field. In FC measurements, a magnetic field of 250 Oe was applied to the samples and then they were cooled from room temperature to 5K and moment data were recorded as the samples cooled.

Figures 4.12 and 4.13 show the M - T curves in both FC and ZFC under at magnetic field of 250 Oe for ZN0 and ZN1 samples, respectively. The magnetic transition temperature (T_c) of the ZnO sample was found to be 280 K. The magnetic transition temperature of the ZN1 sample decreased to 275 K compared to the ZN0 sample.

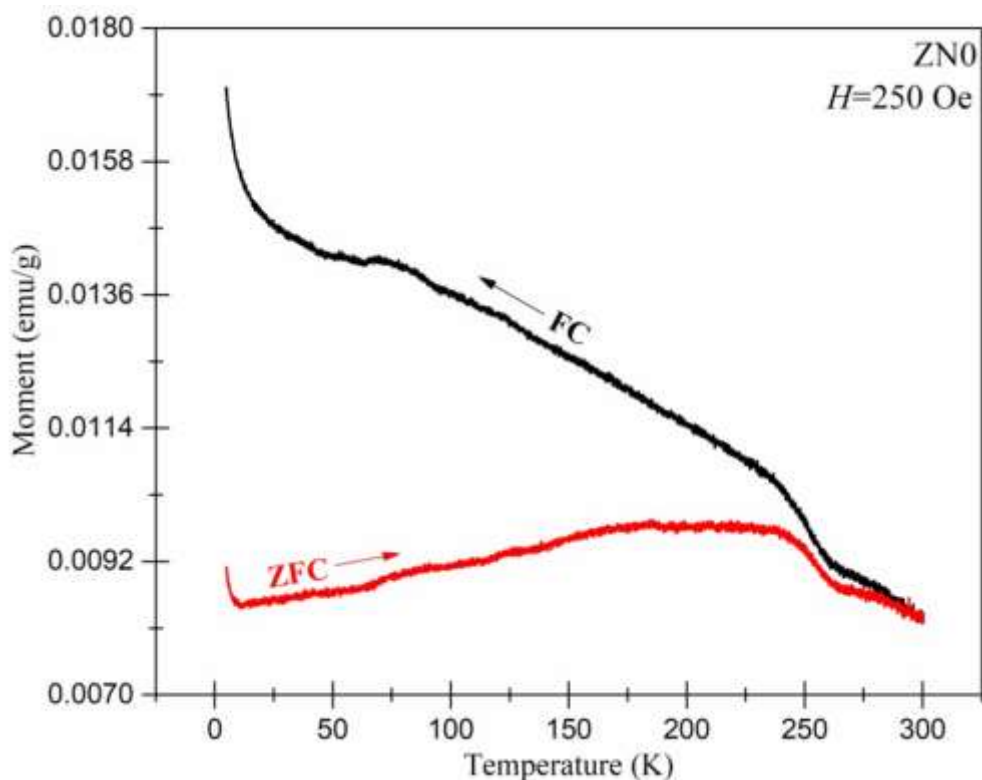


Figure 4.12. Temperature dependent magnetic moment curves of $\text{Ni}_y\text{:ZnO}$ for $y=0$.

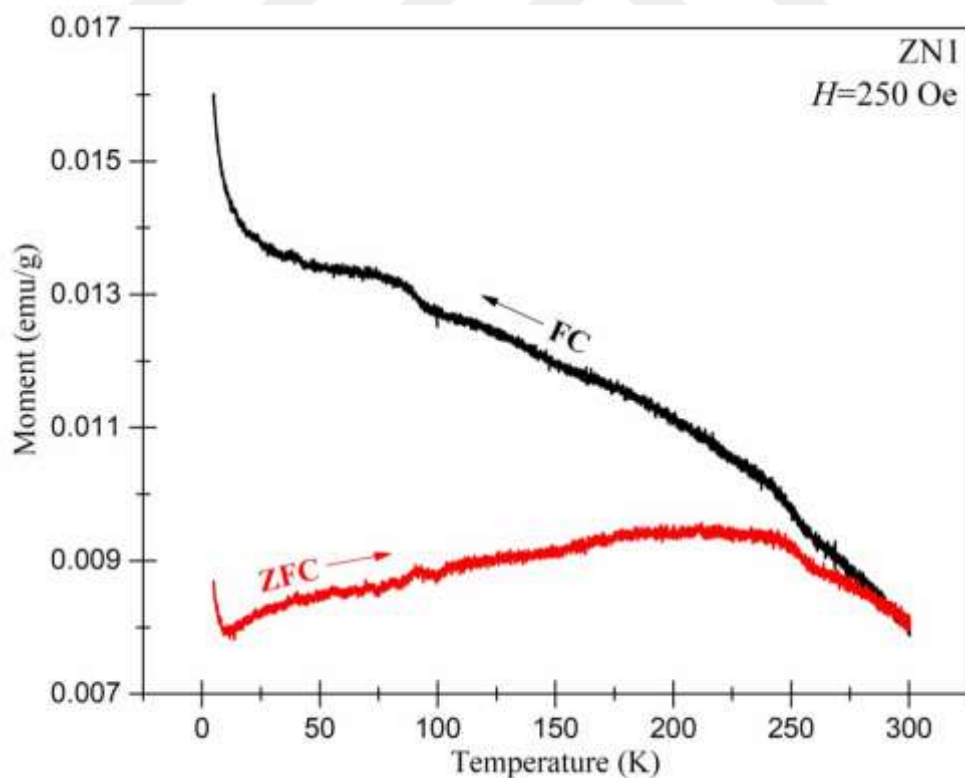


Figure 4.13. Temperature dependent magnetic moment curves of $\text{Ni}_y\text{:ZnO}$ for $y=0.1$.

4.2.2. Magnetic hysteresis of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ and $\text{Ni}_y\text{:ZnO}$ nanoparticles

Magnetic hysteresis loop (M - H) measurements of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ nanoparticles were carried out by sweeping applied magnetic field at room temperature (300K). M - H curves of $\text{Ni}_y\text{:ZnO}$

nanoparticles were obtained at low (5K) and room temperature (300K). Figs. 4.14, 4.15, 4.16, 4.17 and 4.18 show the magnetic hysteresis curve of the CNF0, CNF1, CNF2, CNF3 and CNF4 samples measured at 300K. The saturation magnetization (M_s), remenant magnetization (M_r) and coercive field (H_c) clearly show that the all samples have ferro/feri-magnetic characteristic at room temperature. M_s values are found as 42.08 emu/g, 39.07 emu/g, 31.93 emu/g, 32.07 emu/g and 28.68 emu/g for CNF0, CNF1, CNF2, CNF3 and CNF4, respectively. Also, M_r values are obtained as 20.05 emu/g, 19.49 emu/g, 16.99 emu/g, 17.06 emu/g and 15.07 emu/g for CNF0, CNF1, CNF2, CNF3 and CNF4, respectively. Furthermore, H_c values are determined as 1190 Oe, 1080 Oe, 890 Oe, 530 Oe and 210 Oe for CNF0, CNF1, CNF2, CNF3 and CNF4, respectively. The M_s , M_r and H_c values obtained from the hysteresis curves are tabulated in Table 4.5. The detail analysis on these parameters will be given in Fig. 4.19.

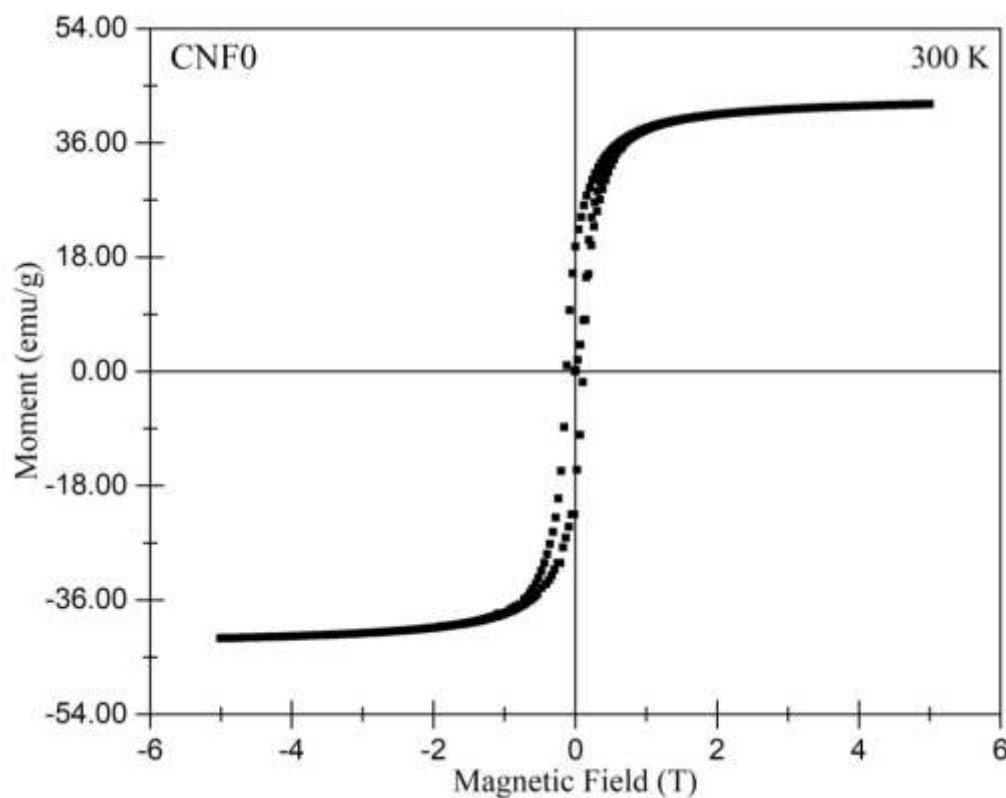


Figure 4.14. Magnetic hysteresis curve of CoFe_2O_4 at 300 K.

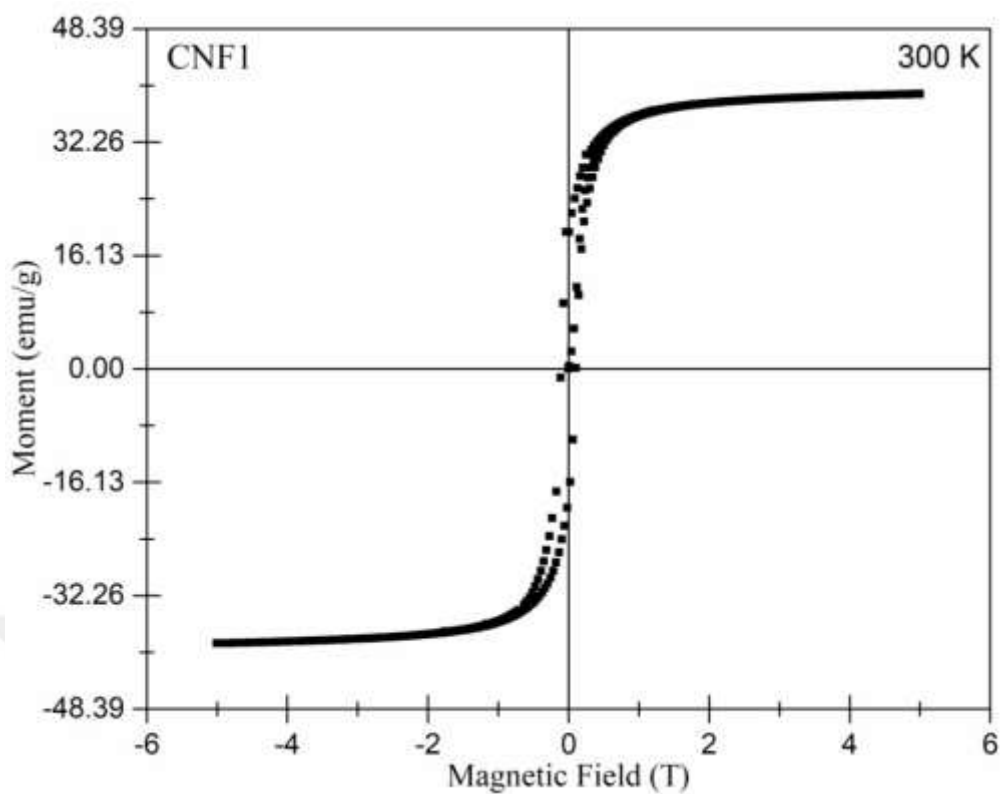


Figure 4.15. Magnetic hysteresis curve of $\text{Ni}_{0.25}\text{Co}_{0.75}\text{Fe}_2\text{O}_4$ at 300 K.

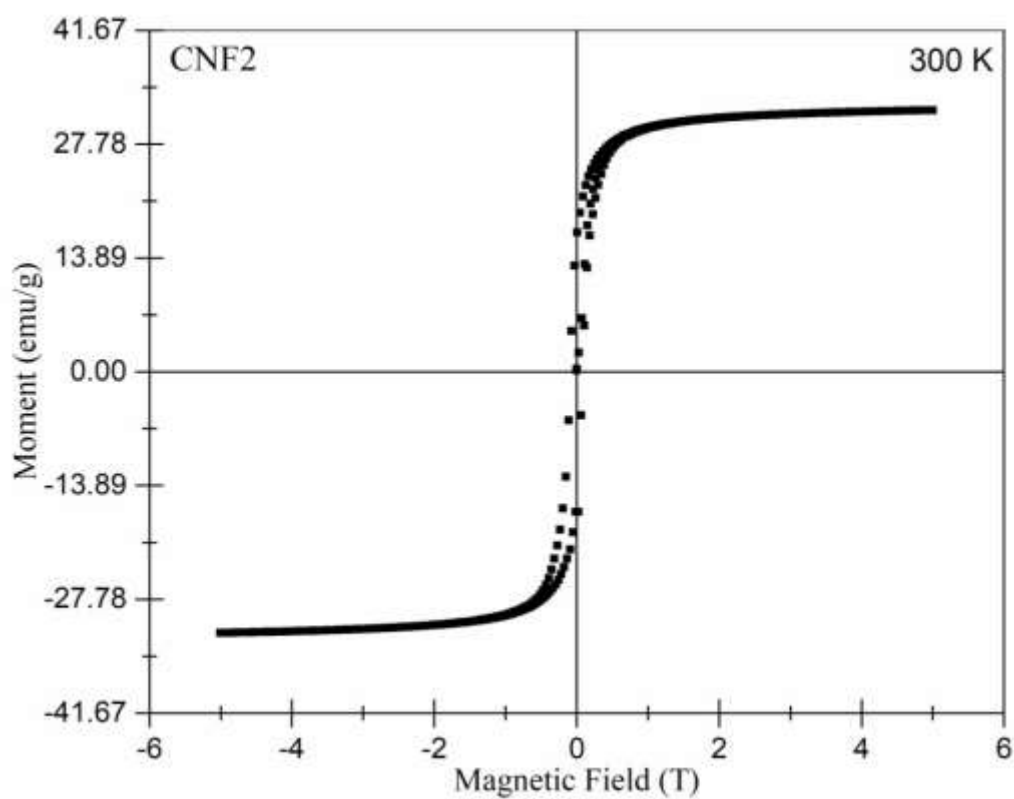


Figure 4.16. Magnetic hysteresis curve of $\text{Ni}_{0.5}\text{Co}_{0.5}\text{Fe}_2\text{O}_4$ at 300 K.

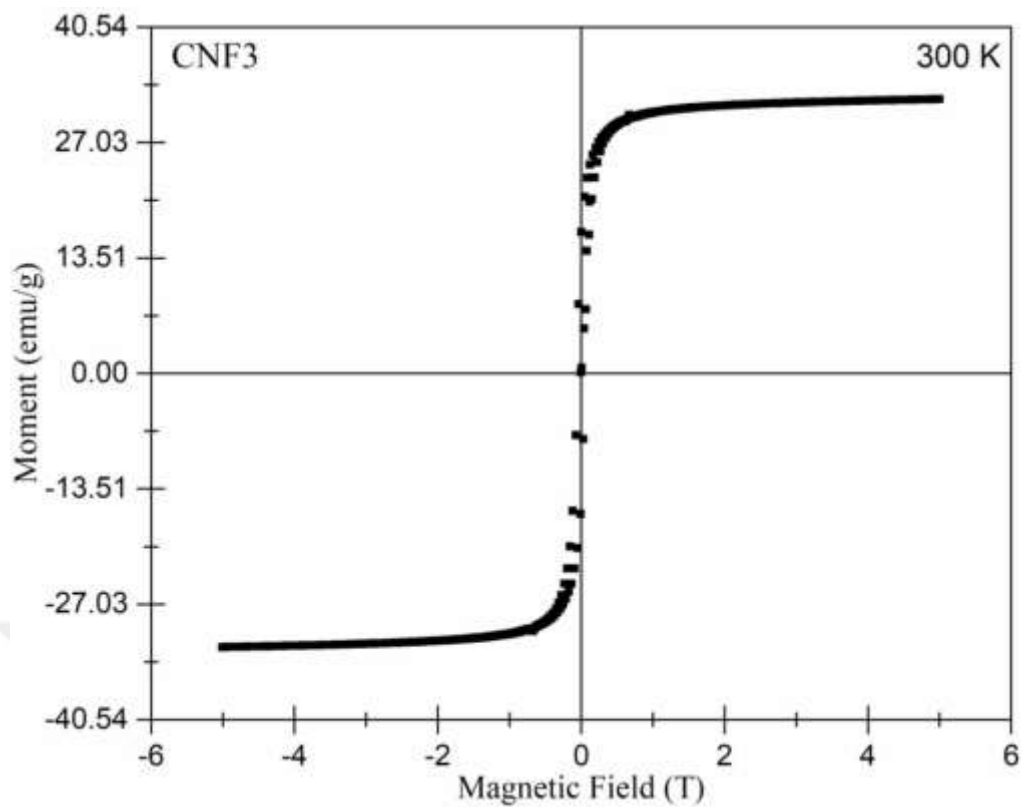


Figure 4.17. Magnetic hysteresis curve of $\text{Ni}_{0.75}\text{Co}_{0.25}\text{Fe}_2\text{O}_4$ at 300 K.

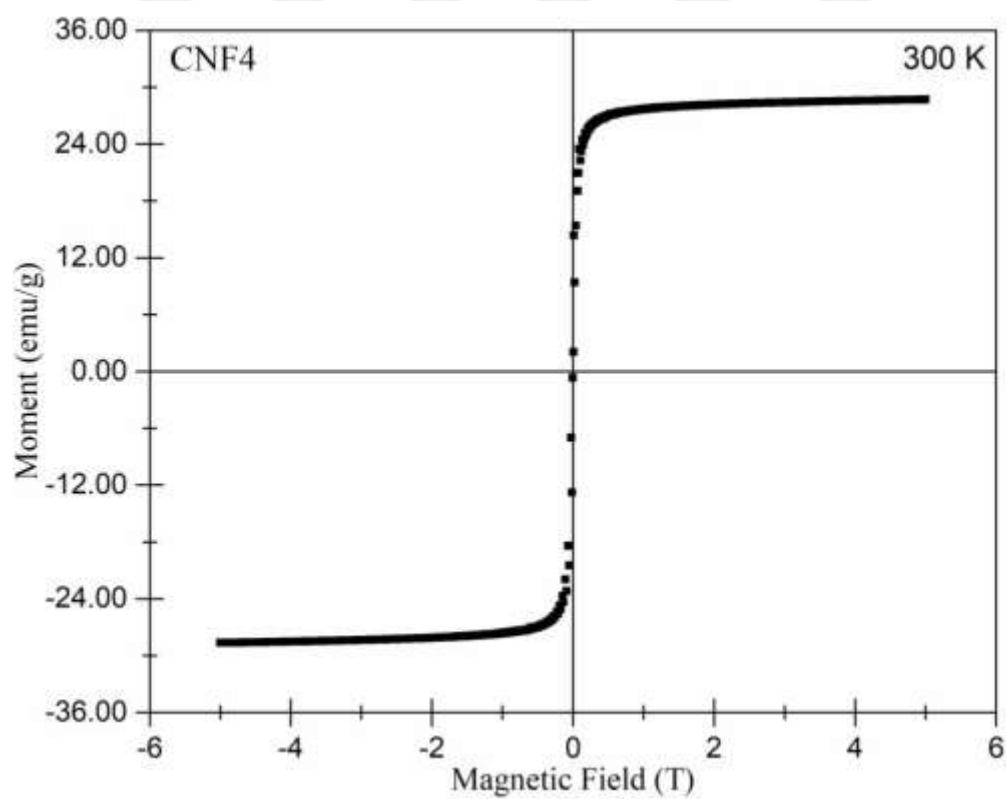


Figure 4.18. Magnetic hysteresis curve of NiFe_2O_4 at 300 K.

Table 4.5. Coercive field (H_c), saturation magnetizations (M_s), remenant magnetization (M_r) and squareness ratio (M_r/M_s) values of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ samples.

Sample Code	H_c (Oe)	M_s (emu/g)	M_r (emu/g)	M_r/M_s
CNF0	1190	42.08	20.05	0.476
CNF1	1080	39.07	19.49	0.499
CNF2	890	31.93	16.99	0.532
CNF3	530	32.07	17.06	0.531
CNF4	210	28.68	15.07	0.525

The H_c , M_s , M_r and M_r/M_s values obtained from hysteresis loops measured at room temperature as a function of Ni content in $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ were given in Figs. 4.19a-d, respectively. The coercive field decreases linearly with increasing amount of nickel as seen in Fig. 4.19a. As it is expected that since the Co (Ni) is a hard (soft) magnetic material, the coercive field decreases with Ni content. According to the Stoner-Wolfforth model ($H_c = \sim 2K/M_s$), the main reason for decreasing in coercivity with increasing Ni content might be related to the magneto crystalline anisotropy that Co^{+2} ion has higher magneto crystalline anisotropy than Ni^{+2} ion. The M_s and M_r decrease with increasing Ni content as shown in Figs. 4.19b-c, respectively. However, the CNF3 sample, Ni content is 75% in main structure, behaves unlike the others and distorts the linear decay. The reason for the increase in M_s and M_r values in CNF3 sample might be related to its highest average particle size as in Fig. 4.9, because the saturation magnetization is directly related to the particle size. The linear decrease in M_s values can be attributed to the magnetic moments of Co^{+2} and Ni^{2+} ions. Since Co^{2+} ions have higher magnetic moment than Ni^{+2} ions, the saturation magnetization decreases when Co^{2+} ions are replaced with Ni^{2+} ions in $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$. Fig. 4.19d gives the M_r/M_s ratio, which is called the squareness ratio. The squareness ratio of the samples increases with Ni amount until the amount of Ni and Co are equal. The maximum M_r/M_s ratio value in the CNF2 sample was found to be 0.532.

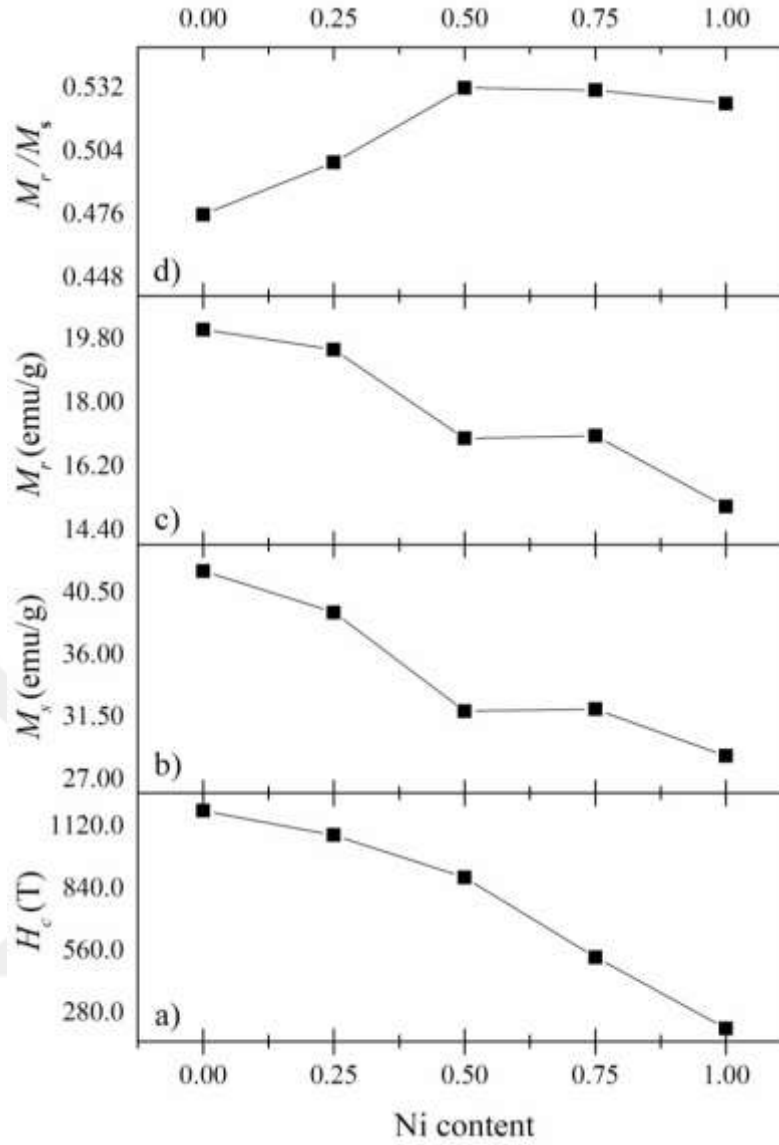


Figure 4.19. a) Coercive field (H_c), b) saturation magnetization (M_s), c) remanence magnetization (M_r) and d) the ratio of squareness (M_r/M_s) of $Ni_xCo_{1-x}Fe_2O_4$ samples at 300K.

Figures 4.20 and 4.21 show the magnetic hysteresis curves of the ZN0 sample measured at 5K and 300K, respectively. The hysteresis loops of the ZN0 samples at 5K indicates that the sample has weak ferromagnetic characteristic having paramagnetic contributions due to a linear increase in magnetization with applied field and not saturated. On the other hand, the ZN0 sample indicates a weak ferromagnetism under low magnetic fields (± 3500 Oe) at 300K. Once the applied field is increased, the diamagnetic contribution becomes dominant as seen in Fig. 4.21. This magnetic behavior of undoped ZnO material is consistent with the literature

(Akyol, Çetin, & Ekicibil, 2016). The M_s , M_r , H_c and M_r/M_s values, were found as 0.182 emu/g (0.032 emu/g), 0.027 emu/g (0.015 emu/g), 520 Oe (340 Oe) and 0.148 (0.469), respectively for 5K (300K), respectively (see Table 4.6).

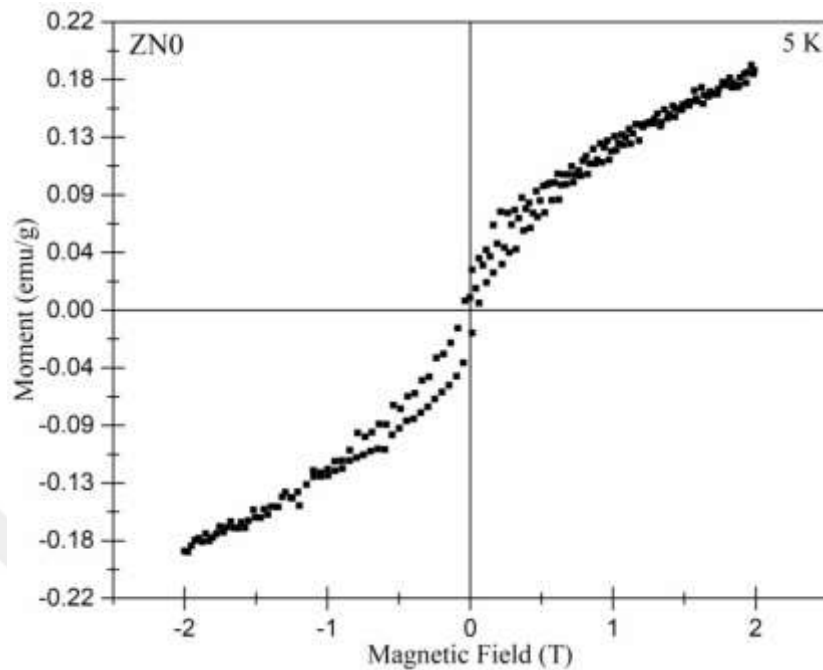


Figure 4.20. Magnetic hysteresis curve of ZnO at 5 K.

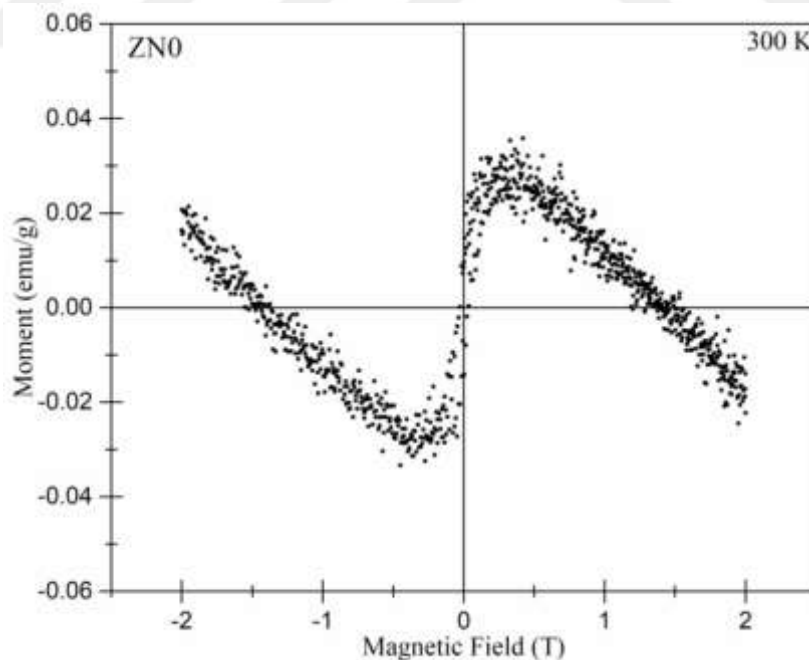


Figure 4.21. Magnetic hysteresis curve of ZnO at 300 K.

Figures 4.22 and 4.23 show the magnetic hysteresis curves of the ZN1 sample measured at 5K and 300K, respectively. The M - H curve measured at 5K indicates that the ZN1 sample has weak ferromagnetic nature. In addition, the positive slope in Figs. 4.22 indicate the presence of

paramagnetic contribution. On the other hand, the sample has negative slope when it is measured at 300K. This negative slope means that the sample has diamagnetic characteristic under high magnetic field region. The M_s , M_r , H_c and M_r/M_s values were found as 0.212 emu/g (0.037 emu/g), 0.024 emu/g (0.017 emu/g), 510 Oe (480 Oe) and 0.113 (0.459), respectively, for 5K (300K) (see Table 4.6).

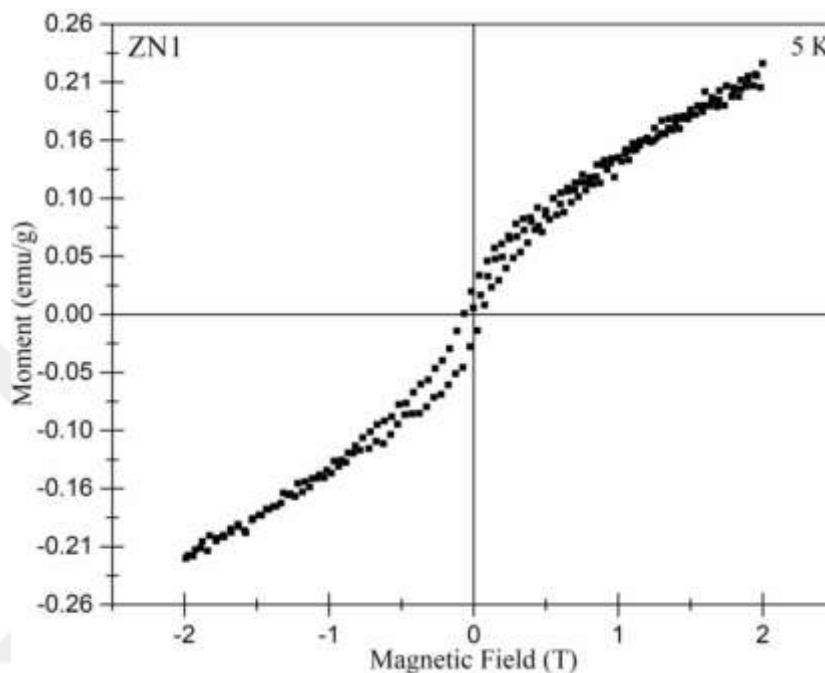


Figure 4.22. Magnetic hysteresis curve of $\text{Ni}_{0.1}\text{ZnO}$ at 5 K.

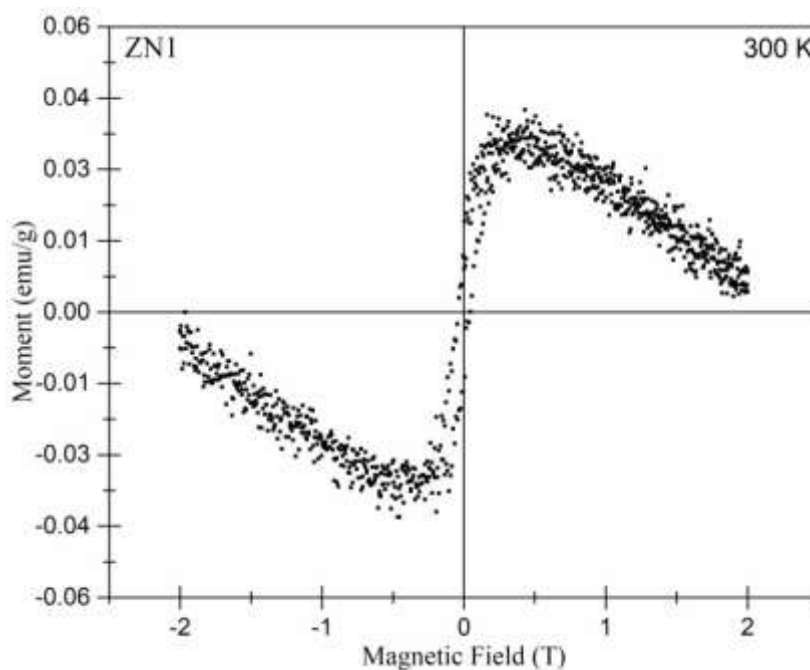


Figure 4.23. Magnetic hysteresis curve of $\text{Ni}_{0.1}\text{ZnO}$ at 300 K.

The magnetic saturation value of ZN1 sample increases when Ni atoms are added to the main structure. Since Ni has ferromagnetic property and ZnO is a magnetically paramagnetic/diamagnetic material, the Ni-doped ZnO sample shows a high magnetization.

Table 4.6. Coercive field (H_c), saturation magnetization (M_s), remenant magnetization (M_r) and squareness ratio (M_r/M_s) values of $Ni_y:ZnO$.

Sample Code	H_c (T)		M_s (emu/g)		M_r (emu/g)		M_r/M_s	
	5K	300K	5K	300K	5K	300K	5K	300K
ZN0	0.052	0.034	0.182	0.032	0.027	0.015	0.148	0.469
ZN1	0.051	0.048	0.212	0.037	0.024	0.017	0.113	0.459

4.3. Microwave absorption properties of $Ni_xCo_{1-x}Fe_2O_4$ and $Ni_y:ZnO$ nanoparticles reinforced epoxy matrix composites

The reflection loss (RL) measurement of various radar absorbing composite structures which were given with their abbreviations in Table 3.1, were measured with the Vector Network Analyzer. RL properties have been measured in the frequency range 8-12 GHz which is the X band. The reflection loss values are specified in decibels (dB) form. The negative increase in the reflection loss plottings indicate that increase the absorbed amount of electromagnetic wave by material. Reflection loss plottings are result of S_{11} parameters versus energy band obtained from waveguides measurement. According to the expressions stated in the literature, the reflection loss value of -10 dB corresponds to 90% absorbance (Akınay, 2018; Bayrakdar, 2011a). -10 dB reflection loss decreases 90% of the power, -20 dB reflection loss decreases 99% of the power.

Figure 4.24 shows the RL versus the frequency graph of a pure epoxy matrix without nanoparticles. In the plotting, a maximum reflection loss value of -11.12 dB was observed at the 11.65 GHz band. The band width value obtained at -10 dB is 0.32 GHz (see Table 4.7).

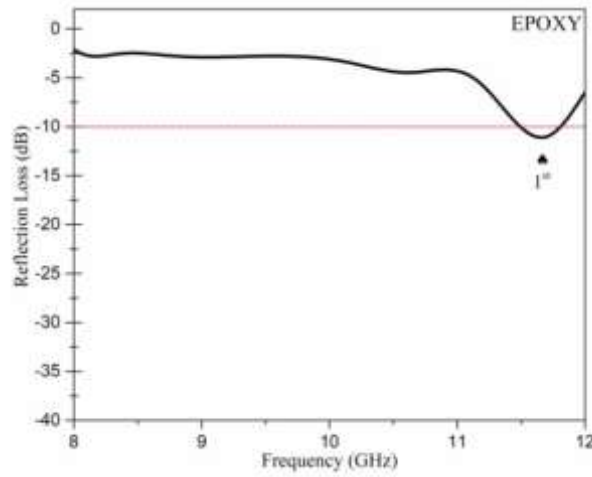


Figure 4.24. Reflection loss plot of the pure epoxy resin without nanoparticles.

Figure 4.25 shows the RL versus the frequency plot of the epoxy matrix reinforced with 15% weight ZnO and 15% weight CoFe_2O_4 nanoparticles called as RAC1 composite. In the figure, a maximum reflection loss value of -20.74 dB was observed at 11.25 GHz. In addition to this peak, a weak absorption is observed at around 9.1 GHz. According to the literature, paraffin/ZnO composite structure gives absorption peak at 9.2 GHz (Wu et al., 2011). Because there is a small difference between paraffin and epoxy in terms of their electromagnetic parameters, the comparison of absorption peak values between the reference (Wu et al., 2011) and our sample would be reasonable (Zhang, Xu, Lin, Yang, & Zhang, 2014). In addition, Zainal et al. (2020) observed the maximum reflection loss value of -29.29 dB at 11.32 GHz for CoFe_2O_4 nanoparticle powder (Zainal, Dedi, & Manaf, 2020). The interaction between dielectric ZnO and magnetic CoFe_2O_4 nanoparticles might lead to little shifting upper and lower frequency sides (the same situation acceptable for RAC7, RAC8 and RAC9). In our case, the absorption peaks observed around 9.1 GHz and 11.25 GHz in RAC1 composite composed of ZnO and, CoFe_2O_4 are consistent with the literature. The band width of the RAC1 composite sample at -10 dB was found to be larger than 1.68 GHz (see Table 4.7). Because we only worked in X-band region, absorption characteristics above 12 GHz could not be obtained.

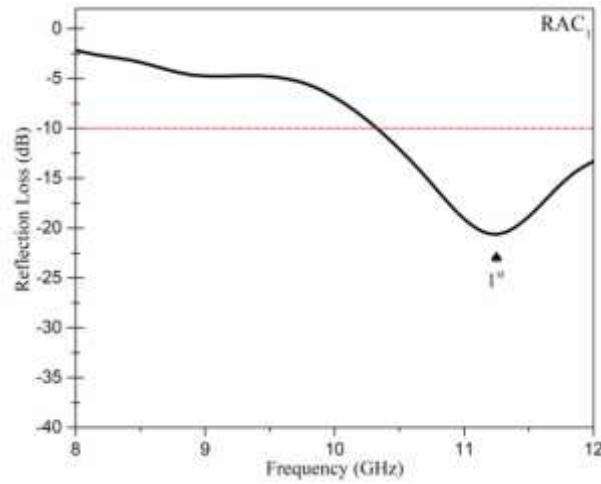


Figure 4.25. Reflection loss plot of the 15% weight ZnO+15% weight CoFe₂O₄ nanoparticles reinforced epoxy RAC1 composite.

Figure 4.26 shows the RL versus frequency plot of the epoxy matrix reinforced with 15% weight Ni_{0.1}ZnO and 15% weight CoFe₂O₄ nanoparticles called as RAC2 composite. As a result of the analysis, two main absorption peaks were observed at 10.66 GHz and 11.79 GHz with maximum reflection loss values of -31.32 dB and -21.72 dB, respectively. As we explained above that CoFe₂O₄ nanoparticle shows absorption at 11.32 GHz (Zainal et al., 2020). This indicates that our absorption peak at 11.79 GHz is corresponding to the CoFe₂O₄. Likewise, the absorption peak at 10.66 GHz for RAC2 composite might be due to the Ni_{0.1}ZnO. Similar absorption peaks were also observed in RAC3 and RAC4 composites. The band widths of these reflections of composite sample at -10 dB were found as 0.87 and 0.70 GHz, respectively (see Table 4.7). Absorption characteristics above 12 GHz could not be obtained because we only worked in the 8-12 GHz (X band) range.

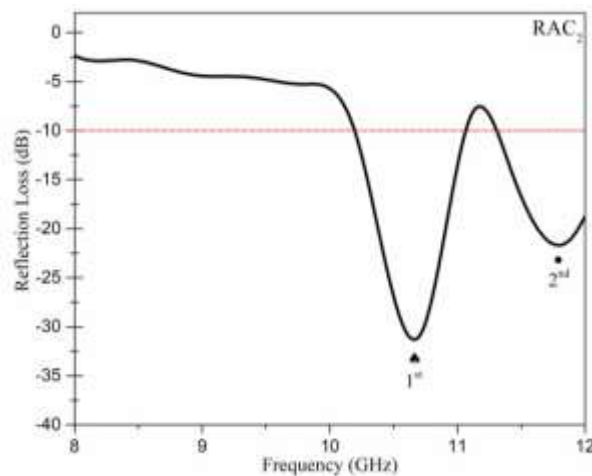


Figure 4.26. Reflection loss plot of the 15% weight Ni_{0.1}ZnO+15% weight CoFe₂O₄ nanoparticles reinforced epoxy RAC2 composite.

Figure 4.27 shows the RL versus frequency plot of the epoxy matrix reinforced with 15% weight $\text{Ni}_{0.1}\text{ZnO}$ and 15% weight $\text{Ni}_{0.25}\text{Co}_{0.75}\text{Fe}_2\text{O}_4$ nanoparticles called as RAC3 composite. As a result of the analysis, two absorption peaks were observed at 10.64 GHz and 11.47 GHz with maximum reflection loss values of -16.75 dB and -15.8 dB, respectively. As explained above that the first (second) absorption peak is corresponding to the $\text{Ni}_{0.1}\text{ZnO}$ ($\text{Ni}_{0.25}\text{Co}_{0.75}\text{Fe}_2\text{O}_4$). The band width of the composite sample at -10 dB was found as 1.70 GHz (see Table 4.7). Absorption characteristics above 12 GHz could not be obtained because we only worked in the 8-12 GHz (X band) range.

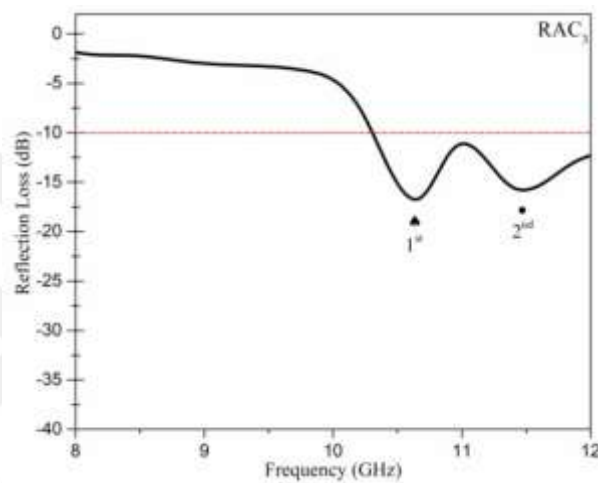


Figure 4.27. Reflection loss plot of the 15% weight $\text{Ni}_{0.1}\text{ZnO}$ +15% weight $\text{Ni}_{0.25}\text{Co}_{0.75}\text{Fe}_2\text{O}_4$ nanoparticles reinforced epoxy RAC3 composite.

Figure 4.28 shows the RL versus frequency plot of the epoxy matrix reinforced with 15% weight $\text{Ni}_{0.1}\text{ZnO}$ and 15% weight $\text{Ni}_{0.5}\text{Co}_{0.5}\text{Fe}_2\text{O}_4$ nanoparticles called as RAC4 composite. As a result of the analysis, two absorption peaks were observed at the 10.64 GHz and 11.60 GHz with maximum reflection loss values of -22.68 dB and -19.69 dB, respectively. As a result of the analysis of composite sample at -10 dB, 0.70 and 0.80 GHz band widths were found for these two absorptions (see Table 4.7). Absorption characteristics above 12 GHz could not be obtained because we only worked in the 8-12 GHz (X band) range.

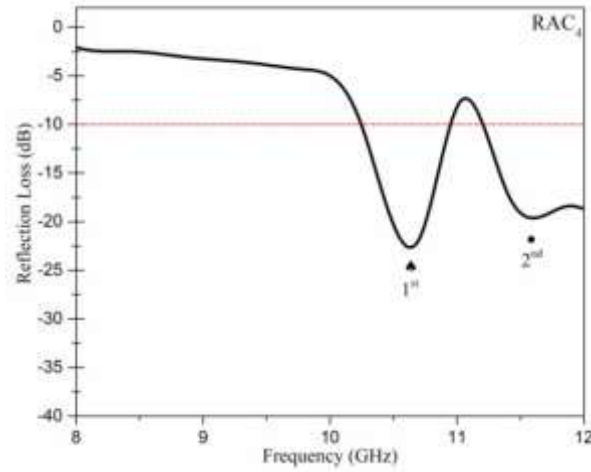


Figure 4.28. Reflection loss plot of the 15% weight $\text{Ni}_{0.1}\text{ZnO}$ +15% weight $\text{Ni}_{0.5}\text{Co}_{0.5}\text{Fe}_2\text{O}_4$ nanoparticles reinforced epoxy RAC4 composite.

Figure 4.29 shows the RL versus frequency plot of the epoxy matrix reinforced with 15% weight $\text{Ni}_{0.1}\text{ZnO}$ and 15% weight $\text{Ni}_{0.75}\text{Co}_{0.25}\text{Fe}_2\text{O}_4$ nanoparticles called as RAC5 composite. As a result of the analysis, two main absorption peaks were observed at 8.4 GHz and 10.4 GHz with reflection loss values of -27.17 dB and -23.80 dB, respectively. The band width of the composite sample at -10 dB was found as 3.07 GHz (see Table 4.7). It would be reasonable to state that the wide absorption band obtained in this structure indicates a good impedance matching via dielectric and magnetic properties of $\text{Ni}_{0.1}\text{ZnO}$ and $\text{Ni}_{0.75}\text{Co}_{0.25}\text{Fe}_2\text{O}_4$ nanoparticles in epoxy matrix. Absorption properties below 8 GHz could not be obtained because the measurements were carried out in the 8-12 GHz (X band) range.

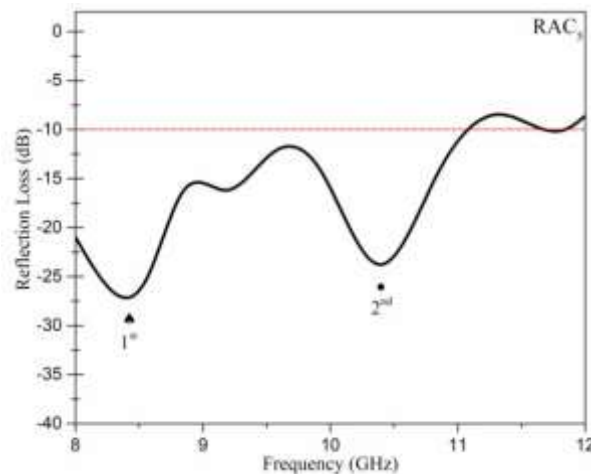


Figure 4.29. Reflection loss plot of the 15% weight $\text{Ni}_{0.1}\text{ZnO}$ +15% weight $\text{Ni}_{0.75}\text{Co}_{0.25}\text{Fe}_2\text{O}_4$ nanoparticles reinforced epoxy RAC5 composite.

Figure 4.30 shows the RL versus frequency plot of the epoxy matrix reinforced with 15% weight $\text{Ni}_{0.1}\text{ZnO}$ and 15% weight NiFe_2O_4 nanoparticles called as RAC6 composite. As a result of the analysis, two absorption peaks were observed at 8.88 GHz and ~ 12 GHz with reflection loss values of -10.85 dB and -19.83 dB, respectively. According to the literature, NiFe_2O_4 /epoxy composite structure showed 11.2 GHz absorption peak for 4 mm thickness structure (Akinay, Hayat, Kanbur, Gokkaya, & Polat, 2018). Though the highest absorption peak's frequency of RAC6 composite is not detected completely because of the measurement limit, it is obviously seen that the peak shifts to the higher frequency when it is compared with reference. The reason of this situation might be experimental and measurement conditions such as (homogeneity, device, etc.). The band widths of the composite sample analyzed at -10 dB were found as 0.34 and 1.47 GHz, respectively (see Table 4.7). Absorption characteristics above 12 GHz could not be obtained because the measurements were carried out in the 8-12 GHz (X band) range.

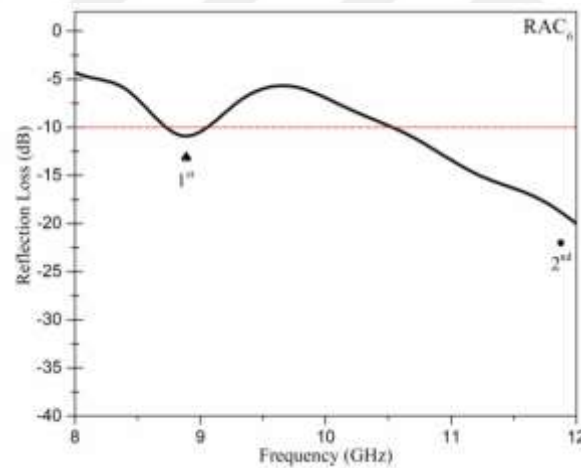


Figure 4.30. Reflection loss plot of the 15% weight $\text{Ni}_{0.1}\text{ZnO}$ +15% weight NiFe_2O_4 nanoparticles reinforced epoxy RAC6 composite.

Figure 4.31 shows the RL versus frequency plot of the epoxy matrix reinforced with 15% weight ZnO and 15% weight $\text{Ni}_{0.25}\text{Co}_{0.75}\text{Fe}_2\text{O}_4$ nanoparticles called as RAC7 composite. As a result of the analysis, a main absorption peak was observed at 10.73 GHz with reflection loss value of -26.08 dB. The band width of the RAC7 composite at -10 dB was found as 1.96 GHz (see Table 4.7).

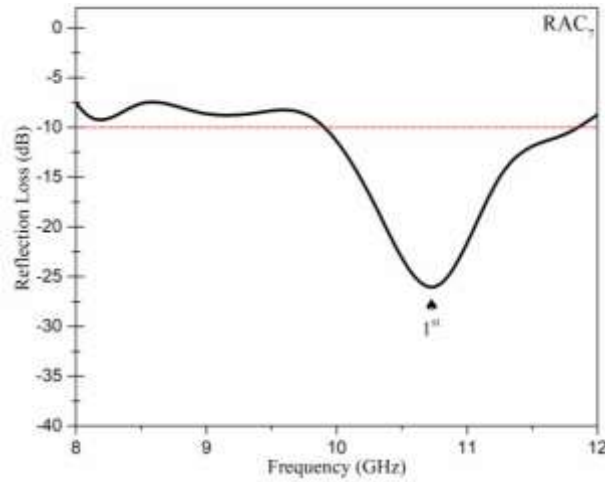


Figure 4.31. Reflection loss plot of the 15% weight ZnO+15% weight $\text{Ni}_{0.25}\text{Co}_{0.75}\text{Fe}_2\text{O}_4$ nanoparticles reinforced epoxy RAC7 composite.

Figure 4.32 shows the RL versus frequency plot of the epoxy matrix reinforced with 15% weight ZnO and 15% weight $\text{Ni}_{0.5}\text{Co}_{0.5}\text{Fe}_2\text{O}_4$ nanoparticles called as RAC8 composite. As a result of the analysis, single absorption band was observed at 11.34 GHz with reflection loss value of -22.98 dB. The band width of the RAC8 composite at -10 dB was found as 2.45 GHz (see Table 4.7). Absorption characteristics above 12 GHz could not be obtained because the measurements were carried out in the 8-12 GHz (X band) range.

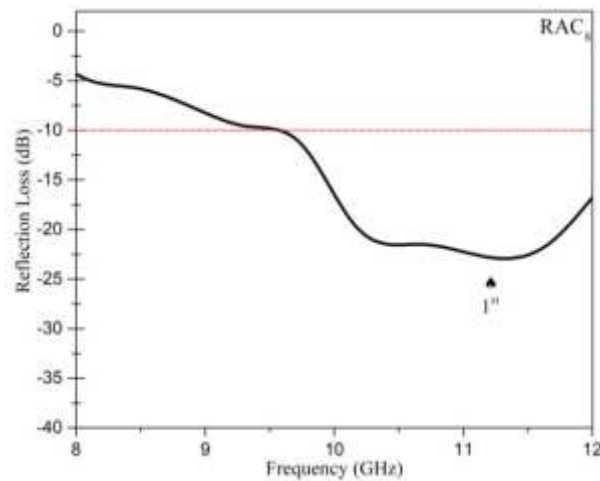


Figure 4.32. Reflection loss plot of the 15% weight ZnO+15% weight $\text{Ni}_{0.5}\text{Co}_{0.5}\text{Fe}_2\text{O}_4$ nanoparticles reinforced epoxy RAC8 composite.

Figure 4.33 shows the RL versus frequency plot of the epoxy matrix reinforced with 15% weight ZnO and 15% weight $\text{Ni}_{0.75}\text{Co}_{0.25}\text{Fe}_2\text{O}_4$ nanoparticles called as RAC9 composite. As a result of the analysis, two absorption bands were observed at 10.23 GHz and 11.39 GHz with reflection loss values of -28.10 dB and -17.79 dB, respectively. The band width of the

composite sample analyzed at -10 dB was found as 2.32 GHz (see Table 4.7). Absorption characteristics above 12 GHz could not be obtained by applying the measurements in the 8-12 GHz (X band) range.

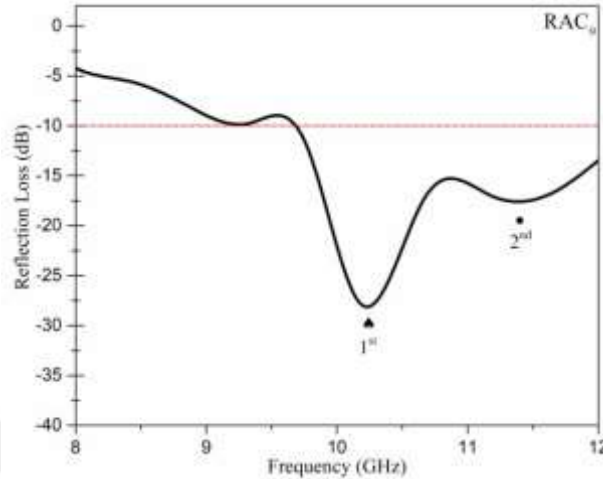


Figure 4.33. Reflection loss plot of the 15% weight ZnO+15% weight $\text{Ni}_{0.75}\text{Co}_{0.25}\text{Fe}_2\text{O}_4$ nanoparticles reinforced epoxy RAC9 composite.

Figure 4.34 shows the RL versus frequency plot of the epoxy matrix reinforced with 15% weight ZnO and 15% weight NiFe_2O_4 nanoparticles called as RAC10 composite. As a result of the analysis, two absorption peaks were observed at 9.3 GHz and 10.68 GHz with reflection loss value of -11.06 dB and -25.35 dB, respectively. According to the mentioned ZnO and NiFe_2O_4 reference samples, 9.2 GHz and 11.2 GHz absorption peaks were observed, respectively. There is a little frequency band shifting was occurred in RAC10 sample. The main reason of shifting is related with the interaction between ZnO and NiFe_2O_4 . The band width of the RAC10 composite at -10 dB was found as 2.95 GHz (see Table 4.7). Absorption characteristics above 12 GHz could not be obtained because the measurements were carried out in the 8-12 GHz (X band) range.

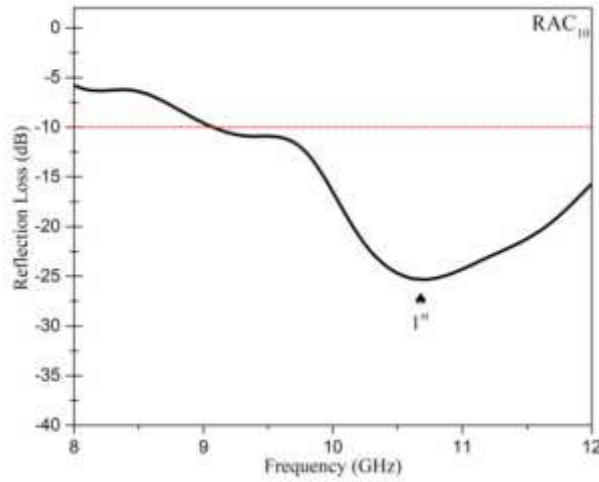


Figure 4.34. Reflection loss plot of the 15% weight ZnO+15% weight NiFe₂O₄ nanoparticles reinforced epoxy RAC10 composite.

Table 4.7 shows the maximum RL at corresponding frequency values and bandwidth at -10 dB of epoxy matrix composite materials combined with various combinations of nanoparticles. As a result of the analysis of the reflection loss properties, it is observed that it has similar reflection loss peaks at similar frequencies by the same additive. The highest reflection loss value was observed at 10.66 GHz with RL -31.32 dB in the RAC2 composite sample. The highest bandwidth of 3.07 GHz was observed in the RAC5 composite sample.

The structural properties, particle size and distribution of Ni_yZnO and Ni_xCo_{1-x}Fe₂O₄ nanoparticles, which are added to the epoxy matrix, affect the permittivity and permeability properties of the samples and change the absorption frequency bands. The magnetic and dielectric additives by various powders reinforced into the epoxy matrix have changed the electromagnetic absorption properties of the composite samples. The unexpected peaks in the results are due to the distribution of the reinforced powder with in the matrix and/or the imperfections between the composite sample and the metal plate.

Table 4.7. The absorption values of composite structures.

Composites	Frequency	Reflection Loss (dB)	Band Width (GHz)
EPOXY	11.65	-11.12	0.32
RAC1	11.25	-20.74	1.68
RAC2	10.66	-31.32	0.87
	11.79	-21.72	0.70
RAC3	10.64	-16.75 -	1.70
	11.47	15.8	
RAC4	10.64	-22.68	0.70
	11.60	-19.69	0.80
RAC5	8.4	-27.17 -	3.07
	10.4	23.8	
RAC6	8.88	-10.85	0.34
	12	-19.83	1.47
RAC7	10.73	-26.08	1.96
RAC8	11.34	-22.98	2.45
RAC9	10.23	-28.10	2.32
	11.39	-17.79	
RAC10	9.3	-11.06	2.95
	10.68	-25.35	

4.4. Mechanical properties of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ and $\text{Ni}_y\text{:ZnO}$ nanoparticles reinforced epoxy matrix radar absorbing composite structures

4.4.1. Charpy impact test

Impact test is an important method used to determine the amount of force absorbed by radar absorber structures when they reach the breaking point to be used in military applications. In this study, Charpy impact test was applied to a 30% nanoparticle reinforced epoxy matrix composite sample prepared in ASTM D256 standards as a representative of all samples. As a result, 0.19 Joule impact energy was recorded on the device. The impact energy value is converted into impact strength by dividing the impact energy by the cross-sectional area of the notched. As a result, an impact strength value of $296.88 \frac{\text{J}}{\text{m}^2}$ was obtained. According to the literature, the charpy izod test of the neat epoxy has $1.3 \frac{\text{J}}{\text{m}^2}$ impact energy (Ergün, 2019). The impact energy value of our composite sample have much larger than the epoxy.

4.4.2. Hardness test

Hardness is important factor that affecting on the abrasion resistance of radar absorbing composite materials which have potential applications in air, land and sea areas. The relative

hardness values are determined by the durometer hardness of rubber, epoxy resin and plastic type materials. In this study, Shore D hardness test was applied to 30% nanoparticles reinforced epoxy matrix composite sample prepared in ASTM D785 standards as a representative of all samples. The hardness values taken from five different regions on the sample are shown in Fig. 4.35. The hardness values of the composite samples in the thesis study were obtained by arithmetic mean of 5 different hardness values taken from various parts of the sample (see Table 4.8). The hardness values taken from 5 different points were close to each other showed a homogeneous distribution of the nanoparticles into the epoxy.

As a result, the average Shore D hardness value was obtained as 62.3 for composite samples.

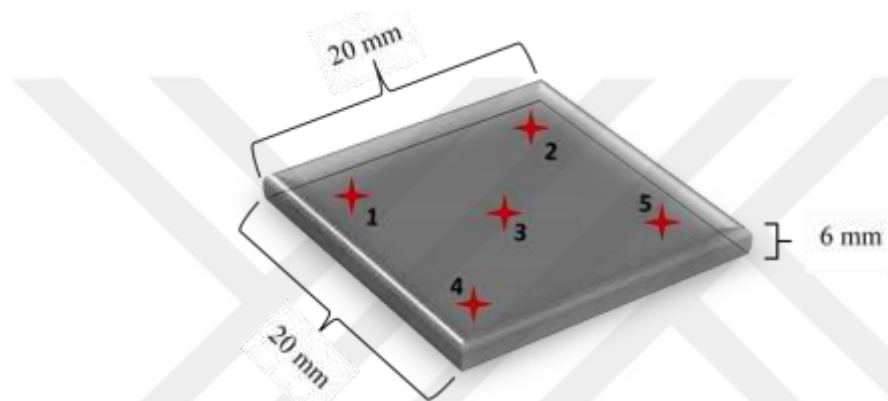


Figure 4.35. Locations of hardness values taken from different places on the sample surface.

Table 4.8. Hardness values taken from five different point on the sample.

Testing Through Various Location	Hardness Value (Shore D)
1	63
2	64
3	61.5
4	61
5	62

5. CONCLUSIONS

In the scope of the thesis, primarily, $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ ($x=0$ to 1, with increments 0.25) and $\text{Ni}_y\text{:ZnO}$ ($y=0$ and 1) nanoparticles were synthesized by sol-gel method and their structural and magnetic properties were investigated. Then, the produced nanoparticles were mixed with epoxy resin and radar absorbing composite structures were obtained. The reflection loss properties and mechanical properties (hardness and impact test) of the radar absorber composite structures were investigated.

All samples were synthesized by sol-gel method. Burning process was applied to the synthesized samples at 500 °C for 1 hour. After burning, the samples have been calcined at 550 °C for 5 hours. Calcined samples were grinded by mechanical and hand grinder for thirty minutes. Cylindrical tablets with a diameter of 10 mm were obtained by applying a pressure of 20 bar to the samples, which were ground with a hydraulic press device. The prepared tablets were sintered in the oven at 800 °C in air for 12 hours. The samples was re-milled to turn into powder form after the sintering process and made ready for measurement.

Structural properties (lattice parameters, phase determination and crystal structure) of the produced nanoparticles were determined by using X-ray diffractometer. Scanning electron microscope (SEM) analysis was used to determine mean particle size distribution and sample surface morphology and EDS analysis was used to determine the elements in the compound. *FC* and *ZFC*, that is mode of *M-T*, measurements were performed by sweeping temperature between 5 and 300 K for $\text{Ni}_y\text{:ZnO}$ samples for magnetic properties. In addition, magnetic hysteresis curves in other words *M-H* measurements were made for all samples at room temperature. In addition, magnetic hysteresis measurement at low temperatures (5K) was applied to $\text{Ni}_y\text{:ZnO}$ samples. Saturation magnetization (M_s), coercive field (H_c) and remenant magnetization (M_r) values were obtained from *M-H* curves and the results were analyzed.

XRD results of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ nanoparticles showed that the samples have cubic crystal structure belonging to the *Fd 3m* space group. A secondary Fe_2O_3 phase was observed in the structure. The reason for the formation of this phase is based on parameters such as the composition of the precursors, the heating/cooling rate and the solution pH. It was observed that the lattice parameter decreased with the increase of Ni content. The reason for this is that

the ionic radius of the Ni^{+2} is smaller than the ionic radius of the Co^{+2} . The SEM images taken at 20 kX and 50 kX magnifications showed that the particles were regionally agglomerated and they were formed as almost spherical. The average particle size was determined with 100 randomly selected particles obtained from SEM images using ImageJ software. Average particle sizes for $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ samples were in the range of 0.155-0.165 μm , while the CNF3 sample was determined around 0.214 μm . The elemental analysis obtained from EDS measurements shows that it is in harmony with the desired weight and atomic percentages in the compounds and that there is no impurity element in the spectrum. The magnetic hysteresis measurements at room temperature revealed that $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ samples are ferri/ferromagnetic. It was observed that saturation magnetization and remanent magnetization decreased linearly with increasing Ni amount, but CNF3 sample disrupted this decrease. The reason for this is due to the increase in the average particle size.

XRD results of $\text{Ni}_y\text{:ZnO}$ nanoparticles indicated characteristic ZnO diffraction peaks which is a hexagonal crystal structure with $P6_3mc$ space group. In addition to ZnO diffraction peaks, NiO diffraction peaks having cubic crystal structure with $Fm-3m$ were observed in $\text{Ni}_{0.1}\text{:ZnO}$. It was observed that the lattice parameter decreased with the increase of Ni content. The reason for this is that the ionic radius of the Ni^{+2} ion is smaller than the ionic radius of the Zn^{+2} ion. The SEM images taken at 20 kX and 50 kX magnifications showed regionally agglomerated particles which are formed in spherical. The average particle size was determined with 100 randomly selected particles obtained from SEM images using ImageJ software. Average particle sizes of ZnO and $\text{Ni}_{0.1}\text{:ZnO}$ samples were found as 0.234 μm and 0.177 μm , respectively. Particle size decreased with adding Ni into the ZnO structure. The elemental analysis obtained as a result of the EDS measurements showed that it is in harmony with the desired weight and atomic percentages in the compounds and that there is no impurity elements in the spectrum. The transition temperature from weak ferromagnetic to paramagnetic phase was determined from $M-T$ curves. The magnetic transition values of the ZN0 and ZN1 samples were found to be 280K and 275K, respectively. The magnetic hysteresis measurements revealed that $\text{Ni}_y\text{:ZnO}$ ($y=0$ and 0.1) samples have similar trend, but the saturation magnetization increased with adding Ni into the ZnO.

The produced $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ and $\text{Ni}_y\text{:ZnO}$ nanoparticles were reinforced in 30% epoxy resin by weight. A mixer was used to distribute the nanoparticles homogeneously in the epoxy matrix and they were kept in a vacuum chamber for fifteen minutes to prevent air bubbles.

The epoxy matrix radar absorbent composite structures given in Table 3.1, were prepared in silicon mold having 22.86x10.16x4 mm³ dimensions. Microwave absorption properties of radar absorber composite structures were measured with the Vector Network Analyzer in the frequency range of 8-12 GHz. According to the reflection loss data, the highest absorption was determined as -31.32 dB at a frequency of 10.66 GHz in the RAC2 sample. The RAC9 sample followed this value as -28.10 dB at a frequency of 10.23 GHz. The highest bandwidth was obtained in the RAC5 sample as 3.07 GHz. RAC10 sample followed this value with 2.95 GHz bandwidth. The results show that the absorption band frequencies are adjustable with the magnetic and dielectric properties of the substitution and reinforcement elements. In general, it has been observed that the reflection loss values have good and useful results in all samples.

Finally, the hardness and impact results of the reference composite structures produced for mechanical tests were determined as 62.3 Shore D and 296.88 $\frac{m^2}{m^2}$, respectively. It was observed that the mechanical strength of the composites increased when it was compared to pure epoxy.

6. RECOMMENDATIONS

In this thesis, the structural and magnetic properties of $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ ($x=0.0, 0.25, 0.50, 0.75, 1.0$) and $\text{Ni}_y\text{:ZnO}$ ($y=0.0$ and 0.1) nanoparticles produced by sol-gel method were investigated. Microwave absorption and mechanical properties by forming radar absorbing composite materials obtained with various nanoparticles added into epoxy were investigated in this thesis study. The composite structures composed of various dielectric and magnetic material showed satisfactory microwave absorption properties. In this thesis, it is aimed to increase the absorption band gap of radar absorbing composite materials, which is an important research subject in the field of defense industry. For these reasons and taking into account the gains we have achieved, the works that can be applied in the near future are listed below.

- The $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ and $\text{Ni}_y\text{:ZnO}$ reinforced epoxy composite materials can be formed at various thicknesses.
- Single-layer $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ or $\text{Ni}_y\text{:ZnO}$ reinforced epoxy composites can be formed separately and multilayer structures can be formed by adding reinforcements with high absorption properties such as carbon black.
- New composite materials can be produced by adding materials with different $\text{Ni}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$ and $\text{Ni}_y\text{:ZnO}$ ratios into epoxy or different polymer materials.
- It can be reinforced with various reinforcement elements to increase the mechanical strength of composite materials.
- Magnetic, dielectric and electromagnetic properties of CoFe_2O_4 and ZnO compounds should be investigated by using different substituents instead of Ni ions.

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