

Tuning the Electromagnetic Response of Co_2Y Hexaferrites through Ni–Zn Substitution and Graphite Addition

Esmail Kiani

es_kiani@yahoo.com

Malek Ashtar University of Technology

Mohammad Hosein Shams

Malek Ashtar University of Technology

Elham Samadpour

Malek Ashtar University of Technology

Research Article

Keywords: Y-type hexaferrite, Ni–Zn substitution, magnetic resonance, Gilbert damping, reflection loss.

Posted Date: October 22nd, 2026

DOI: <https://doi.org/10.21203/rs.3.rs-11128444/v1>

License:   This work is licensed under a Creative Commons Attribution 4.0 International License.

[Read Full License](#)

Additional Declarations: No competing interests reported.

Tuning the Electromagnetic Response of Co₂Y Hexaferrites through Ni–Zn Substitution and Graphite Addition

Esmail Kiani¹, Mohammad Hosein Shams, Elham Samadpour

College of Electroceramics & Electrical Engineering Malek Ashtar University of Technology,
Iran

Abstract

Tuning the magnetic resonance while maintaining sufficient dielectric loss is important for improving microwave absorption in Y-type hexaferrites. In this study, the effects of simultaneous Ni–Zn substitution and graphite addition were investigated in Ba₂Co_{2-x}Ni_{x/2}Zn_{x/2}Fe₁₂O₂₂ ($x = 0.0 - 2.0$). The powders were synthesized by the citrate sol–gel method and characterized using X-ray diffraction, scanning electron microscopy, and magnetometry. Complex permittivity and permeability were measured over 1–18 GHz for paraffin composites containing 80 wt% filler, and the reflection loss (RL) was calculated using transmission-line theory. The coercive field decreased from 651 to 45 Oe with increasing x , while the magnetic resonance frequency showed a nonmonotonic variation. The permeability spectra were well described by a distributed Kittel–Gilbert model with a lognormal resonance-frequency distribution. The fitted median resonance frequency increased from 3.04 GHz at $x = 0$ to 10.08 GHz at $x = 0.5$ and then decreased to 2.57 GHz at $x = 2$. The $x = 2$ composite showed an effective absorption bandwidth (EAB; $RL \leq -10$ dB) of 3.7 GHz at a thickness of 4.5 mm. Adding 8 wt% graphite relative to the filler mass increased the EAB to 4.0 GHz at a reduced thickness of 3.0 mm, with a minimum RL of -38 dB at 5.1 GHz and 4.0 mm. Overall, the results show that Ni–Zn substitution can effectively tune the magnetic resonance, while a controlled amount of graphite can improve impedance matching and absorption at lower thicknesses.

Keywords

Y-type hexaferrite; Ni–Zn substitution; magnetic resonance; Gilbert damping; reflection loss.

1. Introduction

Electromagnetic wave-absorbing materials have attracted considerable attention because of their potential to mitigate electromagnetic interference and unwanted electromagnetic radiation while supporting electromagnetic shielding and information-security applications. Y-type hexaferrites are particularly attractive because of their favorable frequency dispersion of magnetic permeability and potential for microwave and high-frequency applications [1-6]. Many Co-containing Y-type compositions exhibit ferroplana behavior at room temperature, in which the basal plane perpendicular to the crystallographic c -axis is the preferred plane of magnetization [7,8]. This planar anisotropy facilitates magnetization rotation within the basal plane and can be associated with relatively low coercivity, high initial permeability, and resonance frequencies that can be tuned through chemical substitution. The effects of cation substitution on the electromagnetic properties of Y-type hexaferrites have been extensively investigated [9-16]. Substitution with cations having different magnetic moments and site preferences can modify the magnetic moments of individual sublattices and alter the superexchange interactions. Among the divalent cations, Co²⁺ has a particularly important effect on the anisotropy of Y-type ferrites.

Corresponding author.¹
E-mail addresses: es_kiani@yahoo.com (E. Kiani).

Replacing Co^{2+} with cations having different electronic configurations and magnetic moments can therefore modify both the anisotropy energy and the magnetic exchange interactions. Ni^{2+} is commonly reported to favor octahedral coordination [10], whereas nonmagnetic Zn^{2+} often shows a preference for tetrahedral sites [17]. However, the actual cation distribution depends on the synthesis conditions and cannot be uniquely determined without direct site-sensitive measurements.

To the best of our knowledge, the electromagnetic response of the $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ substitution series has not been systematically investigated. In the present system, the simultaneous replacement of Co^{2+} by Ni^{2+} and Zn^{2+} preserves charge neutrality but alters the magnetic contributions of the sublattices. This substitution provides a route to investigate how changes in composition shift the magnetic resonance response within the 1–18 GHz frequency range and affect microwave absorption. In particular, the relationship between magnetic softening, the distribution of resonance frequencies, and absorption performance remains to be established for this substitution series.

Graphite was introduced to compensate for the weak dielectric loss of the ferrite and to examine whether the absorption bandwidth could be increased at a reduced thickness. However, an excessive carbon content may produce a highly conductive network, leading to impedance mismatch and increased reflection at the absorber surface. Therefore, the carbon content must be carefully optimized to achieve a suitable balance between wave penetration into the material and attenuation within the absorber.

In this work, Y-type hexaferrite powders with the composition $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ ($x = 0, 0.5, 1, 1.5$, and 2) were synthesized by the citrate sol–gel method. The relationships among phase composition, particle morphology, static magnetic properties, and complex permeability were investigated. The permeability spectra measured over 1–18 GHz were analyzed using a distributed Kittel–Gilbert model, and the microwave absorption of the composites was evaluated using transmission-line theory. The $x = 2$ composition was selected for graphite modification because it combined a substantial effective absorption bandwidth with a lower corresponding absorber thickness than the other compositions. Because the cation distribution was not directly determined in this study, the observed trends are discussed as structure–property correlations supported by XRD, SEM, magnetization, and electromagnetic measurements.

2. Materials and methods

2.1. Synthesis procedure

To synthesize Y-type barium hexaferrite powders with the nominal composition $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ ($x = 0, 0.5, 1, 1.5$, and 2), analytical-grade metal nitrates purchased from Merck were used. Homogeneous 0.2 mol L^{-1} aqueous solutions of the metal nitrates, including $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (>98%), $\text{Ba}(\text{NO}_3)_2$ (>99%), $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (>98%), $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (>98%), and $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (>99%), were prepared separately in distilled water. Appropriate amounts of the nitrate solutions were mixed at 40°C according to the nominal stoichiometry of each composition. The mixture was then heated to $60\text{--}65^\circ\text{C}$ under magnetic stirring. A 0.2 mol L^{-1} citric acid (CA) solution was prepared and then added dropwise as a chelating agent during the sol–gel process, maintaining a molar ratio of CA to $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ of 4:3. Ammonia was subsequently added dropwise to adjust the pH to 7–8, which was selected to promote homogeneous gel formation and subsequent phase development [8]. The sol was maintained at $60\text{--}65^\circ\text{C}$ and pH 7–8 for approximately 15 h to allow water evaporation and gel formation. It was then dried in an oven at 80°C for 48 h. The dried gel was subsequently ground into a fine powder and calcined in an electric furnace under composition-dependent temperature and

time conditions selected to promote the formation of the Y-type hexaferrite phase. The main stages of the citrate sol–gel synthesis process, from the formation of metal–citrate complexes to the development of the crystalline Y-type hexaferrite phase, are schematically illustrated in Fig. 1.

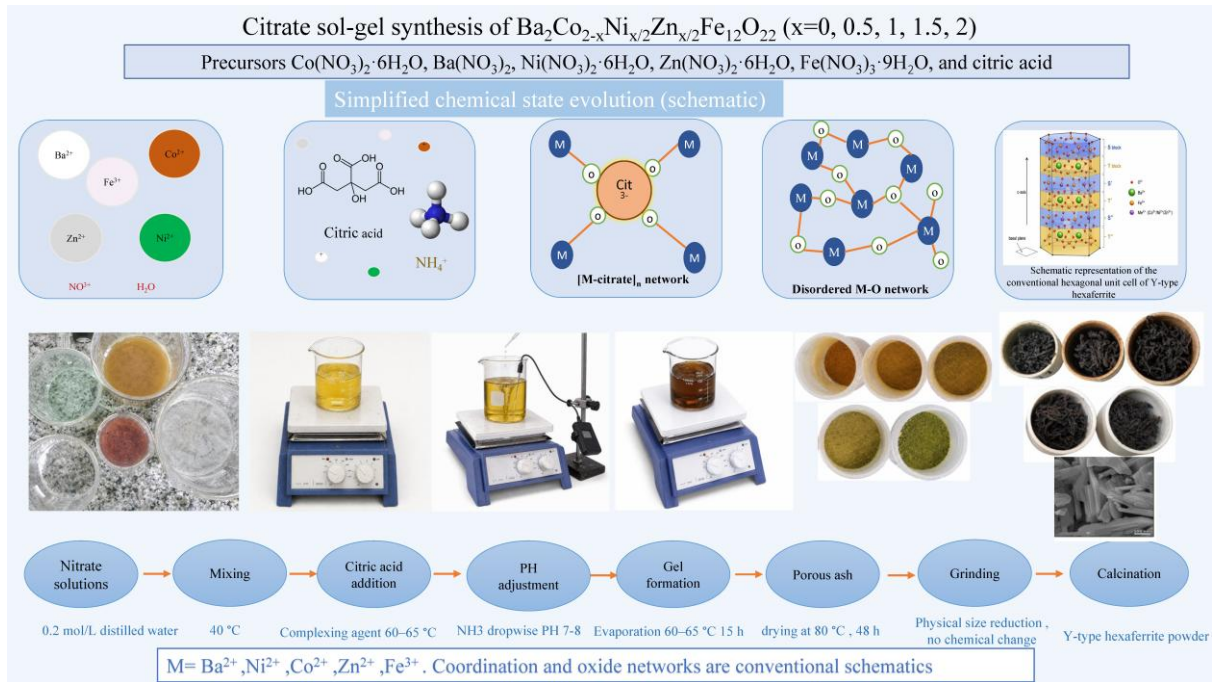


Fig. 1. Schematic representation of the citrate sol–gel synthesis of $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ Y-type hexaferrites.

2.2. Characterization

X-ray diffraction (XRD) analysis was performed to assess phase formation under the investigated calcination conditions. The XRD patterns were recorded using a PANalytical X'Pert Pro MPD using a diffractometer with a Cu anode and an X-ray wavelength of 1.541 Å, over a 2θ range of 10° – 70° . A step size of 0.05° and a counting time of 1 s per step were used at 25 °C. Rietveld refinement of the XRD patterns was performed using MAUD software (Material Analysis Using Diffraction) to investigate the structural properties of the samples. The morphology and microstructure of the calcined hexaferrite particles were examined using a MIRA3 scanning electron microscope (SEM) operating at an accelerating voltage of 15 kV. The static magnetic properties were measured at room temperature using a vibrating sample magnetometer (VSM; Meghnatis Kavir Kashan Co., Iran) under a maximum applied magnetic field of 10 kOe. To investigate the electromagnetic properties, ferrite–paraffin composites containing 80 wt% $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ hexaferrite ($x = 0, 0.5, 1.0, 1.5$, and 2.0) and 20 wt% paraffin was prepared. The mixtures were molded into toroidal specimens with an inner diameter of 3.04 mm, an outer diameter of 7.0 mm, and a thickness of 1–2 mm. The specimens were placed in a coaxial transmission airline connected to an Agilent vector network analyzer (HP 8722ES). The reflection and transmission scattering parameters, S_{11} and S_{21} , respectively, were measured over the frequency range of 1–18 GHz. The complex relative permittivity ($\epsilon_r = \epsilon' - i\epsilon''$) and permeability ($\mu_r = \mu' - i\mu''$) were subsequently extracted from the measured scattering parameters using Agilent material measurement software based on the Nicolson–Ross reflection/transmission formulation.

3. Results and Discussion

3.1. Structural characterization

Fig. 2 presents the X-ray diffraction patterns of the $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ samples ($x = 0, 0.5, 1, 1.5$, and 2) calcined at 950°C for 3 h . The patterns show reflections corresponding to the Y-type hexaferrite phase, together with secondary phases identified as CoFe_2O_4 , BaFe_2O_4 , $\alpha\text{-Fe}_2\text{O}_3$, and M-type $\text{BaFe}_{12}\text{O}_{19}$. With increasing x , the reflections of the secondary phases become progressively more prominent, particularly for $x = 2$, indicating that the Y-type phase is not fully formed under this calcination condition.

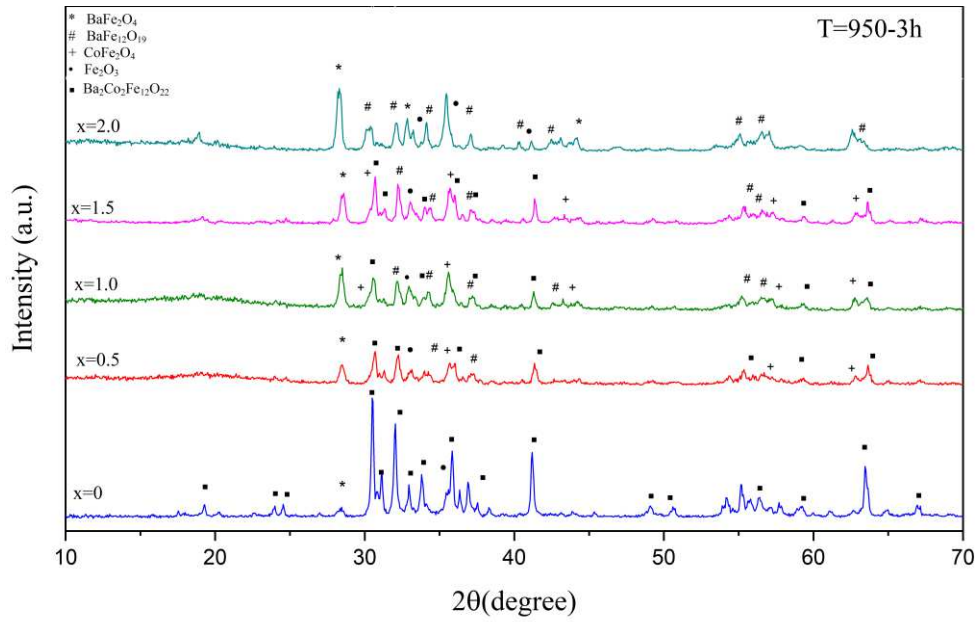


Fig. 2. X-ray diffraction patterns of $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ ($x = 0, 0.5, 1, 1.5$, and 2) calcined at 950°C for 3 h .

In general, Y-type hexaferrite forms indirectly through reactions in the $\text{BaO-Fe}_2\text{O}_3\text{-MeO}$ system, where Me represents a divalent cation [7]. The persistence of secondary phases at higher x suggests that Ni–Zn substitution may retard Y-type phase development under the investigated conditions, so that a higher calcination temperature is required for Y-type phase formation. A contrasting trend was observed in our previous study of Cu–Ni-substituted Y-type hexaferrites, in which the temperature required to obtain a single Y-type phase decreased with increasing x ; this behavior may partly be attributed to the lower melting point and greater high-temperature reactivity of CuO relative to ZnO , which can facilitate cation diffusion and phase formation [9]. Fig. 3 shows the X-ray diffraction patterns of the $x = 0.5, 1$, and 1.5 samples calcined at 975°C for 1 h . Reflections of the Y-type phase are observed in all three samples, together with clearly detectable secondary phases, including CoFe_2O_4 , BaFe_2O_4 , $\alpha\text{-Fe}_2\text{O}_3$, and M-type $\text{BaFe}_{12}\text{O}_{19}$. Despite the shorter dwell time, the Y-type reflections are more pronounced than those obtained at 950°C for 3 h . This comparison indicates that increasing the temperature from 950 to 975°C promotes formation of the desired phase under the present conditions; however, the individual effects of temperature and dwell time cannot be separated quantitatively because both variables were changed.

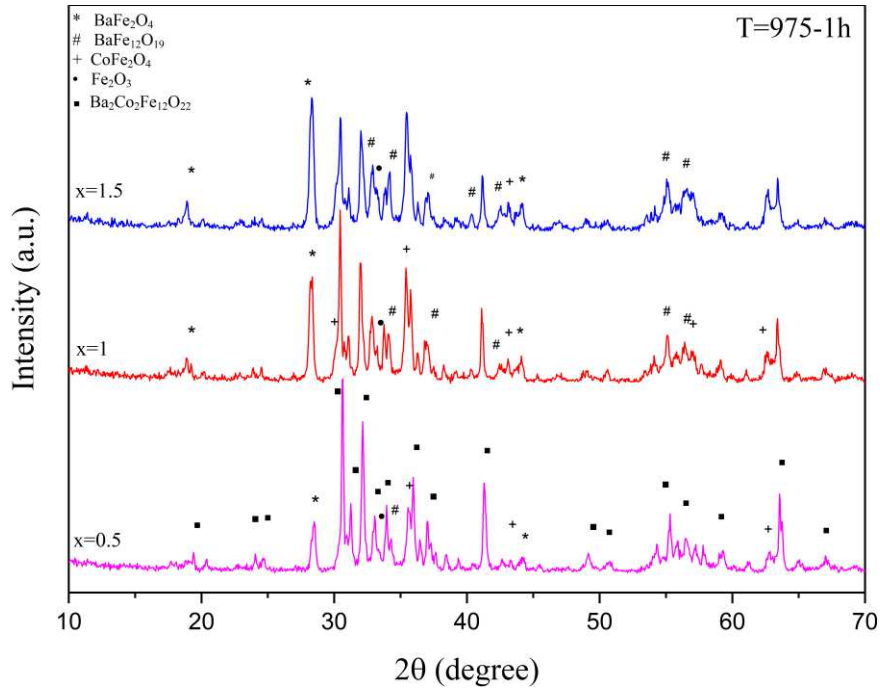


Fig. 3. X-ray diffraction patterns of $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ ($x = 0.5, 1, \text{ and } 1.5$) calcined at 975°C for 1 h.

Based on these preliminary calcination trials, the samples were subsequently calcined at 1000°C for 3 h to promote formation of the Y-type phase. Because secondary phases persisted in the $x = 2$ sample at 1000°C , this composition was also calcined at 1100°C for 3 h. The corresponding XRD patterns are presented in Fig. 4, together with the $x = 0$ pattern obtained at 950°C for comparison. The Miller indices of the principal Y-type reflections are indicated above the corresponding peaks.

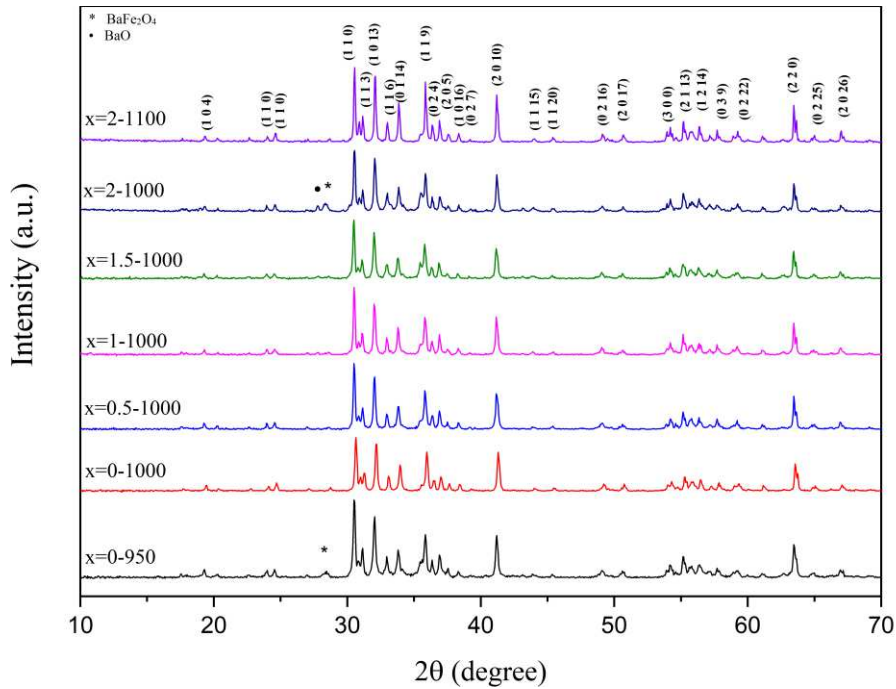


Fig. 4. X-ray diffraction patterns of $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ ($x = 0, 0.5, 1, 1.5, \text{ and } 2$) calcined at 1000°C for 3 h, together with the patterns of $x = 0$ calcined at 950°C and $x = 2$ calcined at 1100°C .

As shown in Fig. 4, the Y-type hexagonal phase is dominant in all samples calcined at 1000°C . No secondary-phase reflections are detected for $x = 0, 0.5, \text{ and } 1$ within the detection limit of the measurement. In contrast, weak reflections assigned to BaFe_2O_4 and BaO appear for $x = 1.5$ and become more pronounced for $x = 2$. No

separate crystalline Ni or Zn containing phases are detected. Although the absence of such reflections does not by itself establish the cation distribution, it is consistent with incorporation of the substituted cations into the dominant Y-type structure. For $x = 2$, increasing the calcination temperature to 1100 °C eliminates the detectable secondary-phase reflections, yielding a single Y-type phase within the detection limit of XRD. Relative to the pattern obtained at 1000 °C, the Y-type reflections exhibit narrower peaks. The peak narrowing observed for $x = 2$ is consistent with the increase in crystallite size from 68.53 nm at 1000 °C to 91.37 nm at 1100 °C (Table 1). Calcination temperatures of 950–1100 °C have previously been reported for the formation of sol–gel-derived Y-type hexaferrites [9, 16, 17, 18]. The positions of the principal reflections in the samples identified as single-phase agree well with the standard pattern of Y-type $\text{Ba}_2\text{Co}_2\text{Fe}_{12}\text{O}_{22}$ (JCPDS card No. 00-044-0206), confirming retention of the Y-type crystal structure across the substitution series. Rietveld refinement of the XRD patterns was performed using MAUD software with a fourth-order polynomial background function. The resulting lattice parameters, crystallite sizes, and axial ratios are summarized in Table 1. For the samples calcined at 1000 °C, a varies from 5.847 to 5.872 Å, c from 43.389 to 43.552 Å, and c/a from 7.388 to 7.440. These small, nonmonotonic variations indicate that the average Y-type lattice dimensions are largely retained upon Ni–Zn substitution. The crystallite size ranges from 41 to 68 nm at 1000 °C and increases to 91 nm for $x = 2$ after calcination at 1100 °C. An expanded view of selected reflections is presented in Fig. 5.

Table 1. Lattice parameters (a and c), crystallite size (D), and axial ratio (c/a) of the $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ samples

x	Calc. Temp. (°C)	a	c	D	c/a
	(3 h)	(Å)	(Å)	(nm)	
0.0	1000	5.847	43.478	47	7.435
0.5	1000	5.862	43.519	59	7.423
1.0	1000	5.853	43.552	41	7.440
1.5	1000	5.872	43.389	48	7.388
2.0	1000	5.860	43.539	68	7.429
2.0	1100	5.856	43.548	91	7.436

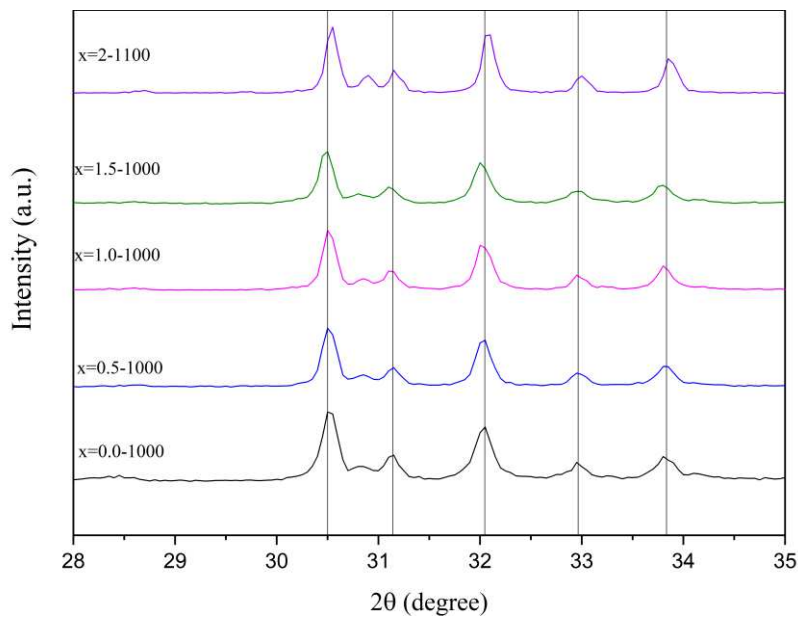


Fig. 5. Expanded view of selected X-ray diffraction reflections of $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$, showing the peak shifts.

The modest changes in the lattice dimensions may arise from the different ionic radii and local coordination preferences of Ni^{2+} , Zn^{2+} , and Co^{2+} , as well as from substitution-induced lattice strain. The simultaneous substitution of Ni^{2+} and Zn^{2+} may partially offset their individual structural effects, resulting in the observed nonmonotonic behavior. Nevertheless, because the cation distribution was not measured directly, specific crystallographic site occupancies cannot be assigned solely from the peak shifts. Moreover, because the $x = 2$ single-phase sample was obtained at a higher calcination temperature than the other compositions, its peak positions and crystallite size cannot be attributed exclusively to composition.

3.2. Morphological analysis

The SEM micrographs of the $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ samples are presented in Fig. 6. All compositions exhibit the characteristic plate-like morphology of Y-type hexaferrites, although the degree of particle growth, stacking, and agglomeration varies with composition. The preferential development of the basal planes is consistent with the anisotropic crystal structure of hexaferrites, in which the growth rates along the in-plane directions and the crystallographic c-axis are different. This anisotropic growth produces thin platelets with relatively large lateral surfaces. Such a morphology is favorable for microwave-absorbing applications because it can modify shape anisotropy, magnetic-domain configuration, interparticle interactions, and the propagation path of electromagnetic waves [10]. The particles are generally arranged as overlapping and interlocked platelets, with local regions showing step-like stacking and partial neck formation. The microstructure does not evolve monotonically with increasing Ni–Zn substitution. The intermediate compositions contain comparatively finer and more elongated platelets, whereas the $x = 2$ sample exhibits pronounced coarsening and noticeably thicker plates. The extensive growth and partial joining of the particles in this sample are most reasonably attributed to its higher calcination temperature, which promotes diffusion, grain-boundary migration, and sintering-assisted particle coalescence.

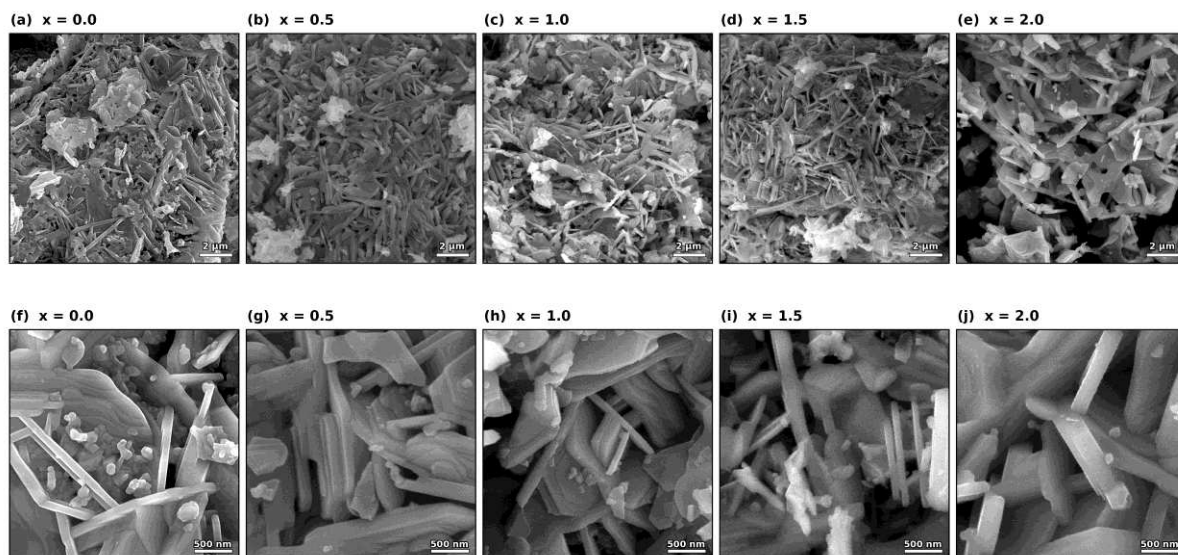


Fig. 6. SEM images of $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$: (a, f) $x = 0$, (b, g) $x = 0.5$, (c, h) $x = 1$, and (d, i) $x = 1.5$ calcined at 1000 °C; (e, j) $x = 2$ calcined at 1100 °C.

3.3 Magnetic properties

The room-temperature magnetic hysteresis loops of $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ are presented in Fig. 7, and the corresponding magnetic parameters are summarized in Table 2. Since some samples still show a slight slope in the high-field region at the maximum applied field of 10 kOe, complete magnetic saturation cannot be

confirmed. Therefore, instead of considering the maximum measured value as the intrinsic saturation magnetization, the magnetization measured at 10 kOe, $M_{10\text{kOe}}$, is reported.

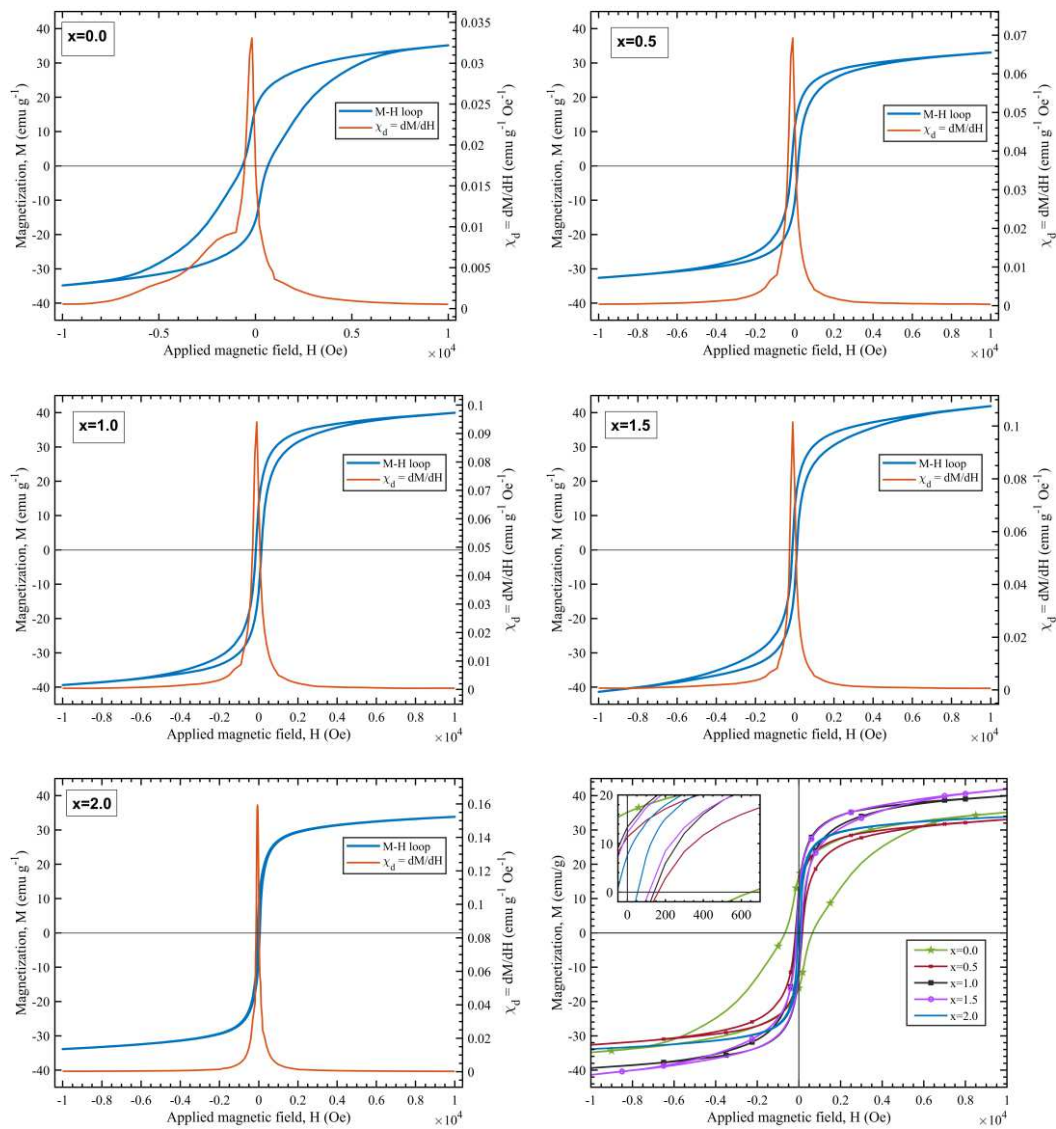


Fig. 7. Room-temperature hysteresis loops and descending-branch differential susceptibility ($\chi_d(H) = dM/dH$) of $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ hexaferrites. The lower-right panel compares the loops, with an inset highlighting the low-field region.

The composition dependence of $M_{10\text{kOe}}$ is nonmonotonic, with a value of 35.14 emu g^{-1} for the unsubstituted Co_2Y sample. The magnetization initially decreases to 33.04 emu g^{-1} at $x = 0.5$, then increases to 39.96 and 41.92 emu g^{-1} at $x = 1.0$ and 1.5 , respectively, before decreasing to 33.82 emu g^{-1} at $x = 2.0$. This behavior cannot be explained solely by the individual magnetic moments of Co^{2+} , Ni^{2+} , and Zn^{2+} . In Y-type hexaferrites, the net magnetization is determined by the difference between the magnetic moments of the antiparallel sublattices located in the S and T blocks. Consequently, the effect of substitution strongly depends on the crystallographic sites occupied by the substituted ions and on the resulting changes in the Fe–O–Fe and Me–O–Fe superexchange interactions [7]. One possible explanation for the increase in magnetization from $x = 0.5$ to $x = 1.5$ is the preferential occupation of spin-down tetrahedral sites by nonmagnetic Zn^{2+} ions, which reduces the moment of the oppositely oriented sublattice. Together with the preferential incorporation of Ni^{2+} into octahedral sites [16], this effect may contribute to the observed increase in magnetization. The decrease at $x = 2.0$ suggests that further substitution changes the balance between the sublattice moments and exchange interactions. However, a definitive interpretation requires direct information about cation-site occupancy, which

could be obtained using techniques such as Mössbauer spectroscopy, X-ray absorption spectroscopy, or neutron diffraction. In addition, because the $x = 2.0$ sample was calcined at a higher temperature and exhibited considerably larger particles, the contributions of compositional and microstructural factors cannot be completely separated.

Table 2. Magnetic parameters of the samples, including the magnetization at 10 kOe ($M_{10\text{kOe}}$), remanent magnetization (M_r), coercive field (H_c), apparent remanence ratio ($S_{10} = M_r / M_{10\text{kOe}}$), the absolute field at the maximum of the descending-branch differential susceptibility ($|H_{\text{peak}}|$), peak differential magnetic susceptibility ($\chi_{d,\text{max}}$), and the full width at half maximum (FWHM) of the differential-susceptibility peak

x	Calcination temperature (3 h)	$M_{10\text{kOe}}$ (emu g ⁻¹)	M_r (emu g ⁻¹)	H_c (Oe)	$S_{10} = M_r / M_{10\text{kOe}}$	$ H_{\text{peak}} $ (Oe)	$\chi_{d,\text{max}}$ (emu g ⁻¹ Oe ⁻¹)	FWHM (Oe)
0.0	1000 °C	35.14	16.42	651	0.467	180	0.033	581.6
0.5	1000 °C	33.04	11.33	162	0.343	100	0.069	619.4
1.0	1000 °C	39.96	13.37	141	0.335	100	0.094	601.1
1.5	1000 °C	41.92	12.26	114	0.293	100	0.101	569.4
2.0	1100 °C	33.82	7.58	45	0.224	60	0.156	189.6

The most pronounced effect of Ni–Zn substitution is observed in the coercive field. The H_c value decreases sharply from 651 Oe for $x = 0$ to 162 Oe for $x = 0.5$ and then continues to decrease to 141, 114, and 45 Oe for $x = 1.0$, 1.5, and 2.0, respectively. Thus, the unsubstituted Co_2Y sample shows the highest resistance to magnetization reversal, whereas the substituted compositions gradually exhibit softer magnetic behavior. This pronounced decrease is consistent with the reduction in Co^{2+} concentration since the strong spin–orbit coupling of Co^{2+} plays an important role in determining magnetocrystalline anisotropy. Particle growth and related changes in grain boundaries, structural defects, internal strain, and domain-wall pinning may also contribute to the reduction in coercivity. In particular, the large plate-like particles observed for $x = 2.0$ are consistent with easier domain-wall motion and the resulting very low coercivity of this sample. However, the present measurements alone do not prove the existence of a multidomain state.

To compare the squareness of the hysteresis loops without assuming complete magnetic saturation, an apparent remanence ratio was defined as $S_{10} = M_r / M_{10\text{kOe}}$. The S_{10} value decreases systematically from 0.467 for $x = 0$ to 0.343, 0.335, 0.293, and 0.224 for $x = 0.5$, 1.0, 1.5, and 2.0, respectively. Therefore, the simultaneous decreases in H_c and S_{10} support the conclusion that replacing Co^{2+} with Ni^{2+} and Zn^{2+} , together with the resulting microstructural evolution, transforms the relatively hard magnetic state of the $x = 0$ composition into a substantially softer magnetic state with increasing substitution. The low-field regions of the magnetic hysteresis loops show a pronounced constriction near the origin, giving the loops a waist-like appearance. Such loops are commonly described as wasp-waisted hysteresis loops [19].

To examine whether this behavior originates from distinct magnetic switching populations, the differential magnetic susceptibility of the descending branch was calculated according to $\chi_d(H) = dM/dH$ [20]. This derivative provides an estimate of the switching-field distribution, in which well-resolved maxima may indicate magnetic populations with distinct reversal fields [20]. The full width at half maximum (FWHM) of each $\chi_d(H)$ peak was also determined as a measure of the breadth of the switching-field distribution. The low-field hysteresis loops and their corresponding differential susceptibility curves are presented in Fig. 7, while the extracted parameters are summarized in Table 2. All compositions exhibit a single stable peak in $\chi_d(H)$, rather than two clearly resolved peaks. Therefore, the observed constriction does not provide evidence for two

independent magnetic phases with well-separated coercive-field distributions, such as distinct soft and hard magnetic phases. Instead, the single peak and its finite FWHM are more consistent with a continuous distribution of local switching and anisotropy fields, in which rapid irreversible reversal at low fields occurs together with a more gradual magnetization-rotation process [20].

With increasing Ni–Zn substitution, the field corresponding to the susceptibility maximum ($|H_{peak}|$) generally shifts toward zero field, in agreement with the overall decrease in coercivity and the apparent remanence ratio, $S_{10} = M_r/M_{10kOe}$. This magnetic softening is most pronounced in the $x = 2$ sample. Given the larger particle size of this sample, its enhanced low-field susceptibility and low coercivity are consistent with a greater contribution from domain nucleation and domain-wall motion. However, randomly oriented platelets with an easy magnetization plane, dipolar interactions, local variations in anisotropy, and domain-wall pinning may also contribute to the broad susceptibility background and the observed wasp-waisted loop shape [20].

3.4. Electromagnetic properties

Fig. 8 shows the frequency spectra of the real (ϵ') and imaginary (ϵ'') parts of the complex permittivity of the $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_x/2\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ hexaferrite composites. Over the frequency range of 1–18 GHz, ϵ' shows relatively weak frequency dispersion for all composites, and no distinct peaks associated with dielectric relaxation or resonance are observed. The highest ϵ' values are obtained for the composite containing the $x = 2.0$ ferrite. This increase may be related to changes in the local lattice polarizability caused by Ni^{2+} and Zn^{2+} substitution, variations in cation distribution, and microstructural factors such as grain growth, reduced porosity, and increased densification resulting from the higher calcination temperature. In contrast, the composites with $x = 0.5$ and $x = 1.5$ exhibit the lowest ϵ' values. The non-linear variation of ϵ' with x indicates that the permittivity is not controlled solely by the substitution level. Cation distribution, lattice defects, microstructure, and the porosity of the composites also contribute to the observed behavior. In ferrite–paraffin composites, the imaginary part of the complex permittivity, ϵ'' , may arise from limited charge-carrier motion, electron hopping between Fe^{2+} and Fe^{3+} ions, ferrite–paraffin interfacial polarization, oxygen-vacancy-related defects, and grain-boundary polarization. However, ϵ'' remains very small for all samples, with an average value below 0.03 over the measured frequency range. This finding indicates that dielectric-loss mechanisms make only a limited contribution to the overall energy dissipation of these composites within the investigated frequency range.

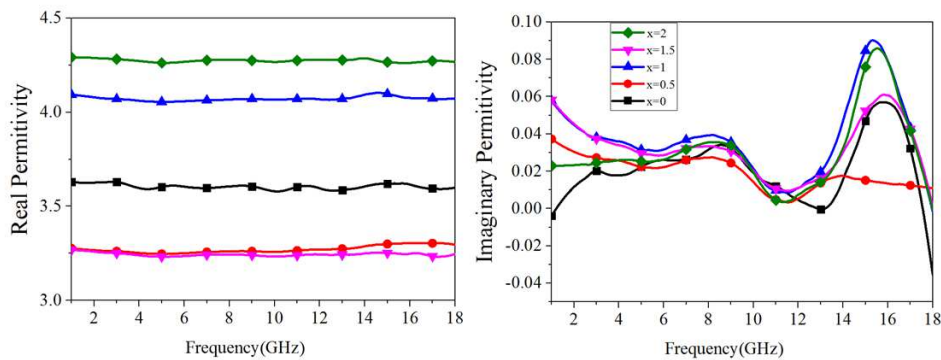


Fig. 8. Frequency dependence of the real part (ϵ') and the imaginary part (ϵ'') of the complex relative permittivity of ferrite–paraffin composites containing 80 wt% $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_x/2\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ ($x = 0.0, 0.5, 1.0, 1.5$, and 2.0).

Fig. 9 shows the frequency dependence of the real (μ') and imaginary (μ'') components of the complex magnetic permeability. For all compositions, μ' exhibits significant dispersion and generally decreases with increasing frequency, indicating that the magnetic moments become less able to follow the rapidly varying alternating

magnetic field. The $x = 2$ sample shows the highest magnetic permeability at low frequencies, whereas the μ' values of all samples approach unity at higher frequencies.

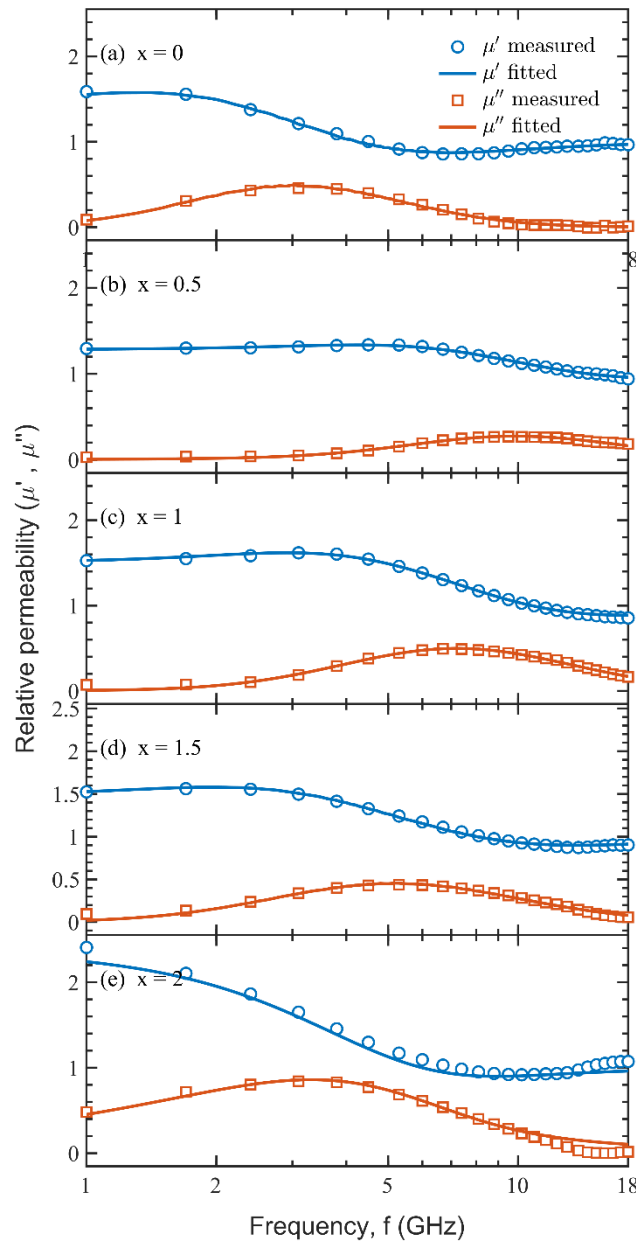


Fig. 9. Measured and fitted frequency-dependent complex magnetic permeability of the ferrite–paraffin composites. Symbols represent the experimental data, and solid lines show the simultaneous fits obtained using the lognormal Kittel–Gilbert model.

The μ'' spectra exhibit broad and asymmetric magnetic-loss peaks at approximately 3, 10, 7, 5, and 3 GHz for $x = 0, 0.5, 1, 1.5$, and 2, respectively. The $x = 2$ sample exhibits the strongest magnetic-loss peak, whereas the $x = 0.5$ sample shows a broader loss band at higher frequencies. The non-monotonic shift of the loss peaks indicates composition-dependent resonant behavior. Furthermore, the broad and asymmetric spectral features are consistent with a distribution of local resonance conditions, rather than a single well-defined resonance frequency. Accordingly, the spectra were analyzed using a distributed Kittel–Gilbert model, as described below. The frequency-dependent magnetic permeability generally arises from two main mechanisms: domain-wall motion and spin rotation or spin resonance. Given the small particle size and the associated suppression of domain-wall motion, the contribution of domain-wall dynamics was assumed to be negligible within the

investigated frequency range [1]. Accordingly, the magnetic response was described using a Kittel-type spin-rotation model with Gilbert damping [21]:

$$\mu^*(f) - 1 = \frac{\chi_{s,0} \left(1 + i\alpha_G \frac{f}{f_r}\right)}{\left(1 + i\alpha_G \frac{f}{f_r}\right)^2 - \left(\frac{f}{f_r}\right)^2} \quad (1)$$

where $\chi_{s,0}$ is the initial susceptibility associated with spin rotation, f_r is the resonance frequency, and α_G is the Gilbert damping parameter. This expression assumes a single well-defined resonance frequency for the entire material. However, the present samples consist of ensembles of particles with different sizes, shapes, orientations, and magnetic anisotropy fields. In addition, dipolar fields arising from interparticle interactions modify the local effective magnetic field. Consequently, individual particles or magnetic regions may exhibit different local resonance frequencies, resulting in a broadened and asymmetric overall response. To account for this inhomogeneous broadening, the formulation of Fiorillo and Beatrice was adapted for the present study to express the total complex permeability as a statistical average of the local resonance responses [22]:

$$\mu^*(f) - 1 = \int_0^\infty P(f_r) \frac{\chi_{s,0} \left(1 + i\alpha_G \frac{f}{f_r}\right)}{\left(1 + i\alpha_G \frac{f}{f_r}\right)^2 - \left(\frac{f}{f_r}\right)^2} df_r \quad (2)$$

The randomly oriented platelets are treated as an effective isotropic medium. Spin rotation is assumed to dominate the magnetic response. A distribution of resonance frequencies is used to represent variations in local resonance conditions. Considering the asymmetric shape of the (μ'') spectra, the distribution of resonance frequencies, $P(f_r)$, was modeled using the following lognormal distribution:

$$P(f_r) = \frac{1}{\sqrt{2\pi} f_r \sigma_{ln}} \exp \left[-\frac{(\ln(f_r/f_m))^2}{2\sigma_{ln}^2} \right] \quad (3)$$

where f_m denotes the median of the resonance-frequency distribution. Accordingly, half of the model distribution lies below f_m . σ_{ln} is the dimensionless parameter describing the degree of inhomogeneous broadening and describes the width of the resonance-frequency distribution on a logarithmic scale. The mean resonance frequency ($\bar{f}_r$) is the expected value, or first statistical moment, of the resonance-frequency distribution and indicates the average position of the distributed local resonances.

$$\bar{f}_r = f_m \exp \left(\frac{\sigma_{ln}^2}{2} \right) \quad (4)$$

The real and imaginary components of the complex magnetic permeability were fitted simultaneously. The measured data and corresponding fitted curves are presented in Fig. 9, while the extracted model parameters and goodness-of-fit indicators are summarized in Table 3. Single-resonance and Gaussian-distribution models were also tested, but both provided poorer fits to the measured permeability spectra than the lognormal-distribution model. The fitted parameters reveal significant and non-monotonic variations in the magnetic dynamics of the samples. The median resonance frequency, f_m , increases from 3.04 GHz for $x = 0$ to a maximum of 10.08 GHz for $x = 0.5$, and then decreases to 7.22, 5.20, and 2.57 GHz for $x = 1$, 1.5, and 2, respectively. The mean resonance frequency follows the same trend, with values of 3.58, 11.44, 8.59, 6.43, and 3.45 GHz, respectively. Meanwhile From $x = 0.5$ to $x = 1.5$, M_{10kOe} increases to 41.92 emu g⁻¹, although the resonance frequency decreases with increasing x beyond $x = 0.5$. This lack of a direct correlation indicates that the resonance

frequency is not determined solely by M_s . Variations in the effective magnetic anisotropy, cation distribution, exchange interactions, and internal magnetic fields must also be considered.

Table 3. Magnetic-resonance parameters and goodness-of-fit indicators obtained using the lognormal Kittel–Gilbert model

x	$\chi_{s,0}$	α_G	$\overline{f_r}$ (GHz)	σ_{ln}	f_m (GHz)	$R_{\mu'}^2$	$R_{\mu''}^2$
0.0	0.45	0.011	3.58	0.57	3.04	0.993	0.990
0.5	0.29	0.177	11.44	0.50	10.08	0.996	0.986
1.0	0.50	0.049	8.59	0.59	7.22	0.997	0.990
1.5	0.48	0.014	6.43	0.65	5.20	0.996	0.981
2.0	1.33	0.22	3.45	0.76	2.57	0.978	0.951

The lowest relative distribution width, σ_{ln} , is obtained for $x = 0.5$ with a value of 0.50, whereas the highest value is observed for $x = 2.0$, reaching 0.76. These values represent the narrowest and broadest relative distributions of local resonance frequencies, respectively. This inhomogeneous broadening cannot be attributed solely to variations in particle size, since differences in crystallographic orientation, local anisotropy, and dipolar interactions may also contribute. For $x = 2$, the considerable increase in the initial susceptibility to 1.33, together with the decrease in H_c to 45 Oe and the reduction in the remanence ratio, M_r/M_{10kOe} , to 0.224, indicates magnetic softening and an increase in the effective magnetic susceptibility. The higher calcination temperature used for this sample, 1100 °C, and the resulting grain growth may also contribute to this behavior. However, the contribution of domain-wall motion is expected to be most significant at frequencies below the measured range. Furthermore, the unusually high damping parameters ($\alpha_G = 0.177$ and 0.22 for $x = 0.5$ and 2.0, respectively), compared with values of 0.011–0.049 for the other samples, should be interpreted as effective damping parameters. In addition to intrinsic Gilbert damping, these effective damping parameters may also reflect overlapping resonance modes and covariance with inhomogeneous broadening. Finally, the R^2 values above 0.95 indicate good numerical agreement over the fitted range.

3.5. Microwave absorption properties

The microwave absorption performance of the 80 wt% $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ –paraffin composites was calculated using the complex permittivity ($\epsilon_r = \epsilon' - i\epsilon''$) and permeability ($\mu_r = \mu' - i\mu''$) measured over 1–18 GHz. For each composition ($x = 0.0, 0.5, 1.0, 1.5$, and 2.0), the normalized input impedance of a metal-backed layer with thickness d was calculated using transmission-line theory [23]

$$\frac{Z_{in}}{Z_0} = \sqrt{\frac{\mu_r}{\epsilon_r}} \tanh \left[i \frac{2\pi f d}{c} \sqrt{\mu_r \epsilon_r} \right] \quad (5)$$

The reflection loss was then obtained from

$$RL = 20 \log_{10} \left| \frac{Z_{in} - Z_0}{Z_{in} + Z_0} \right| \quad (6)$$

Here, Z_{in} is the input impedance of a single-layer absorber, $Z_0 = \sqrt{\frac{\mu_0}{\epsilon_0}}$ is the characteristic impedance of free space, c is the speed of light, f is the frequency of the incident electromagnetic wave and d is the thickness of the absorber. For a metal-backed absorber, an RL value below −10 dB corresponds to more than 90% absorption of the incident electromagnetic energy and is commonly used to define the effective absorption bandwidth. The

absorption minima were analyzed in terms of impedance matching, intrinsic dielectric and magnetic attenuation, and thickness-dependent quarter-wavelength interference. Fig. 10 presents contour maps of the reflection loss (RL) of the $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ -based composites as a function of frequency (1–18 GHz) and absorber thickness (1–10 mm). With increasing Ni–Zn substitution, the minimum RL improves from -11 dB at 4.6 GHz for $x = 0$ to -25 dB at 11.1 GHz for $x = 2$. The RL maps show a non-monotonic dependence on the substitution level. The $x = 0$ composite exhibits the weakest response, with only a narrow region of $RL \leq -10$ dB between 4 and 5 GHz and at thicknesses of approximately 7.5–9.5 mm. For $x = 0.5$, a strong absorption region develops at higher frequencies, and the $RL \leq -20$ dB region extends diagonally from approximately 11 to 18 GHz for thicknesses of 7–10 mm. The $x = 1$ sample maintains this high-frequency response and simultaneously develops a second band with $RL \leq -10$ dB at lower thicknesses of approximately 2.7–4.3 mm, providing greater flexibility in selecting the absorber thickness and operating frequency.

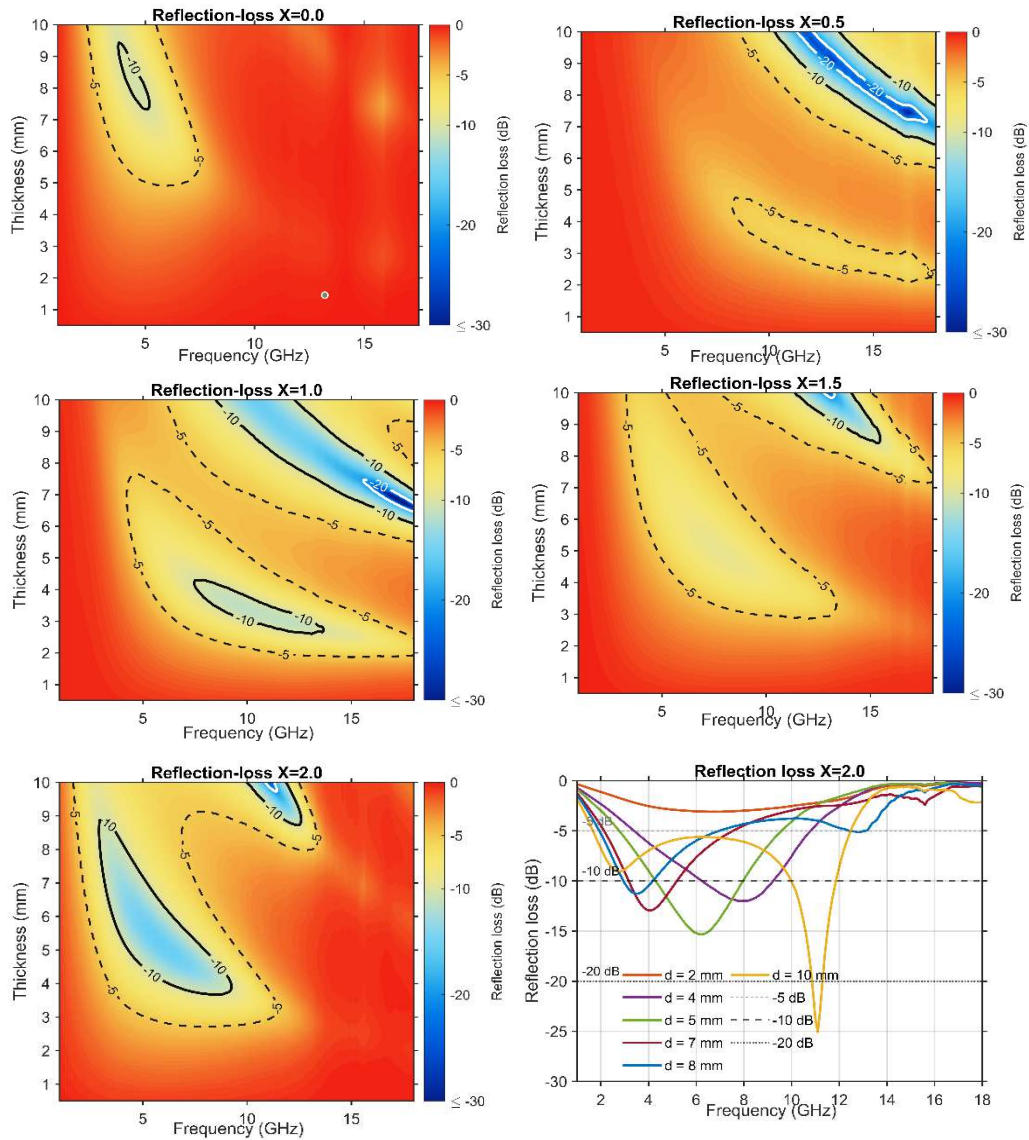


Fig. 10. Reflection-loss (RL) maps of the 80 wt% $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ -paraffin composites ($x=0.0, 0.5, 1.0, 1.5,$ and 2.0) as functions of frequency and absorber thickness for a single-layer, metal-backed configuration. The lower-right panel shows the RL spectra of the $x = 2$ composite at selected thicknesses.

For $x = 1.5$, the absorption performance decreases, and effective absorption is mainly limited to the frequency range of approximately 13–15 GHz and thicknesses above 8.5 mm. For $x = 2$, a broad region with $RL \leq -10$ dB

reappears between approximately 3 and 9 GHz at thicknesses of 4–8.5 mm. In addition, a high-frequency absorption region is observed around 11–13 GHz.

The shift of the absorption minima toward lower frequencies with increasing thickness is consistent with the quarter-wavelength matching condition. In contrast, the composition-dependent changes reflect variations in the balance between impedance matching and dielectric–magnetic attenuation. The microwave absorption characteristics of the $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ composites are summarized in Table 4. These parameters include the minimum reflection loss ($RL_{\min}$), the corresponding frequency ($f_{RL\min}$), maximum effective absorption bandwidth (EAB) defined at $RL \leq -10$ dB, and the absorber thickness corresponding to the maximum EAB (d_{EAB}). The maximum EAB and the global minimum RL were determined independently and may occur at different absorber thicknesses. Their corresponding thicknesses are reported separately.

Efficient microwave absorption requires the incident electromagnetic wave to enter the absorbing medium with minimal reflection, after which various loss mechanisms dissipate its energy as heat. Efficient coupling at the air–absorber interface is favored by good impedance matching between free space ($Z_0 \approx 377 \Omega$) and the ferrite composite. The characteristic impedance of the composite is given by $\eta_m = Z_0 \sqrt{\frac{\mu_r}{\epsilon_r}}$, and the approach of η_m to Z_0 reduces the reflection of the incident wave from the composite surface. Once inside the absorber, the electromagnetic field is attenuated by dielectric and magnetic loss mechanisms. The attenuation constant is related to these parameters through

$$\alpha = \frac{k_0}{\sqrt{2}} \left[\sqrt{(\mu' \epsilon' - \mu'' \epsilon'')^2 + (\mu' \epsilon'' + \mu'' \epsilon')^2} - (\mu' \epsilon' - \mu'' \epsilon'') \right]^{1/2} \quad (7)$$

where $k_0 = 2\pi f/c$ is the free-space wavenumber. In addition to the intrinsic electromagnetic properties, the absorber thickness and operating frequency influence phase accumulation and, consequently, the absorption behavior, as indicated by Eq. (5). Ultimately, for a metal-backed single-layer absorber, reflection is governed by the input impedance of the entire structure, and optimum matching is achieved when Z_{in} approaches Z_0 .

Table 4. Microwave absorption characteristics of $\text{Ba}_2\text{Co}_{2-x}\text{Ni}_{x/2}\text{Zn}_{x/2}\text{Fe}_{12}\text{O}_{22}$ hexaferrite samples for different values of x

Sample (x)	EAB (GHz) for $RL \leq -10$ dB	d_{EAB} (mm)	$f_{RL\min}$ (GHz)	$RL_{\min}$ (dB)	$d_{RL\min}$ (mm)
0.0	0.7	8	4.6	-11	8
0.5	3.8	8	16.7	-28	8
1.0	4.0	8	16.8	-31	7
1.5	2.7	10	13.0	-22	10
2.0	3.7	4.5	11.1	-25	10

The synthesized ferrites exhibit relatively low permittivity together with appreciable magnetic permeability. This combination provides a more favorable balance between ϵ and μ , bringing the characteristic impedance of the material closer to that of free space and facilitating the penetration of electromagnetic waves into the absorber layer. This behavior may provide an advantage over many predominantly dielectric absorbers, which often suffer from impedance mismatch. However, impedance matching alone is not sufficient to achieve effective microwave absorption, as the wave entering the material must also be sufficiently attenuated. In these composites, the attenuation constant is low because of their very small imaginary permittivity, which limits the

attenuation of the incident wave. The frequency-dependent attenuation constant and characteristic impedance of the $x = 2.0$ sample, which provided a substantial EAB at a lower corresponding thickness among the investigated compositions, are presented in Fig. 11.

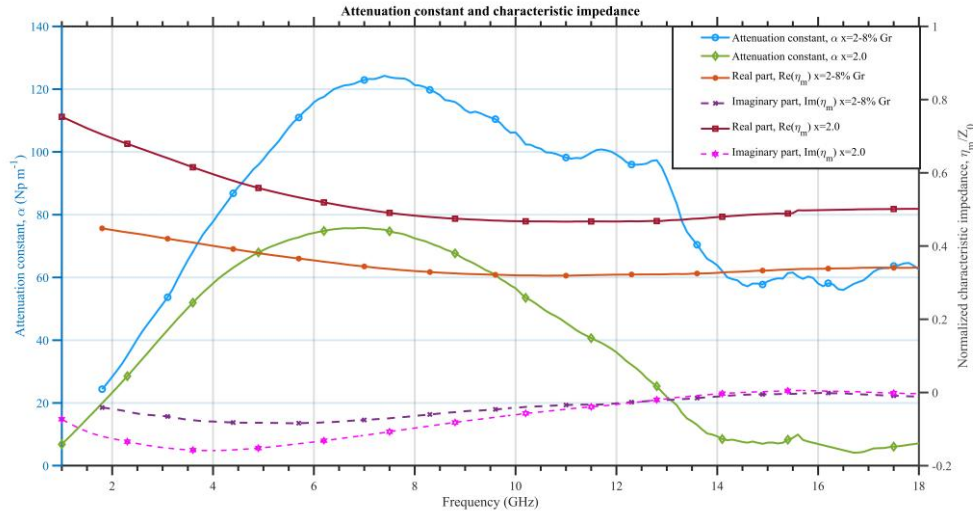


Fig. 11. Frequency dependence of the attenuation constant (α) and the normalized characteristic impedance η_m/Z_0 of the $\text{Ba}_2\text{NiZnFe}_{12}\text{O}_{22}$ /graphite/paraffin composites containing 0 and 8 wt% graphite.

3.6. The effect of adding graphite

Carbon-based materials, including carbon nanotubes and graphite, are widely used to enhance dielectric attenuation in magnetic absorbers. At suitable concentrations, the relatively high electrical conductivity of these materials promotes the formation of conductive pathways within the matrix, thereby increasing both the real and imaginary parts of the complex permittivity. The resulting conduction loss and interfacial polarization can enhance electromagnetic-wave attenuation and improve microwave absorption [24]. Accordingly, graphite was incorporated as a secondary component of the composite to compensate for the relatively low dielectric loss of the ferrite phase. However, an excessive carbon content may produce a highly conductive network, leading to impedance mismatch and increased reflection at the absorber surface. Therefore, the carbon content must be carefully optimized to achieve a suitable balance between wave penetration into the material and attenuation within the absorber.

The $x = 2.0$ composition was selected for graphite modification because it provided an EAB of 3.7 GHz at a thickness of 4.5 mm, whereas the slightly broader bandwidths of the $x = 0.5$ and $x = 1.0$ composites required a thickness of 8 mm. Graphite was added at 8, 15, and 25 wt% relative to the total ferrite–graphite filler mass. For characterization using a vector network analyzer, each hybrid filler was mixed with paraffin to prepare composites containing 80 wt% ferrite–graphite filler and 20 wt% paraffin. The corresponding electromagnetic parameters (ϵ_r , μ_r) of $\text{Ba}_2\text{NiZnFe}_{12}\text{O}_{22}$ graphite–paraffin composites containing 80 wt% total filler are presented in Figs. 12 and 13. As shown in Fig. 12, the real part of the complex magnetic permeability gradually decreases with increasing graphite content. This trend is mainly attributed to the dilution of the magnetically active hexaferrite phase by graphite, whose relative permeability is close to unity. Consequently, the reduction in the ferrite fraction decreases the overall magnetic response of the composite. In addition, as the graphite content increases, the μ'' resonance peak shifts toward higher frequencies, its peak magnitude decreases, and the resonance curves broaden. Since the composition and crystal structure of the ferrite phase remain unchanged, this small shift is more likely related to interfacial and composite effects, such as changes in the magnetic

coupling between particles, local demagnetizing fields, and the electromagnetic environment created by graphite, rather than to a fundamental change in the intrinsic magnetic anisotropy of the hexaferrite.

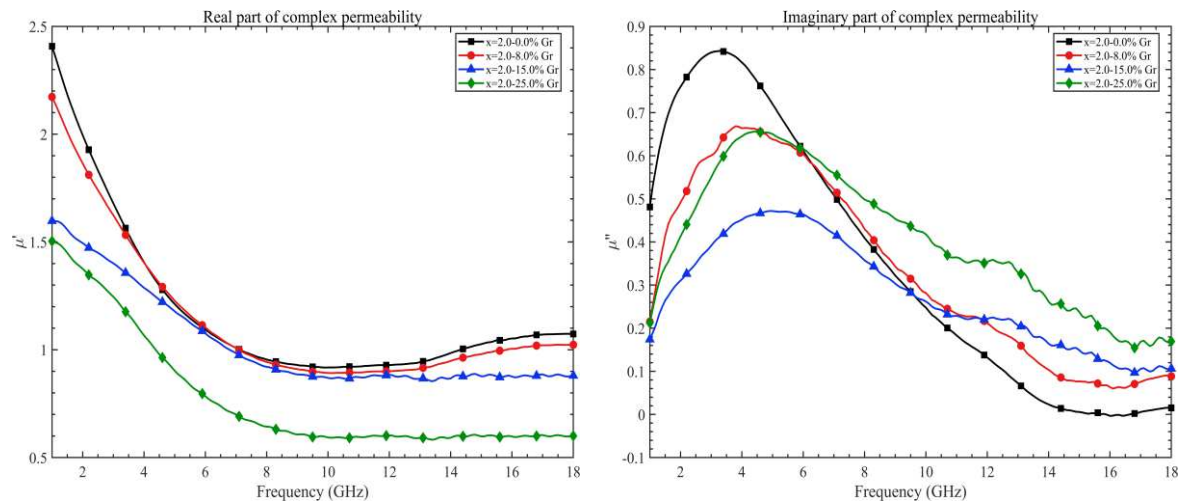


Fig. 12. Frequency dependence of the real part (μ') and the imaginary part (μ'') of the complex relative permeability of $\text{Ba}_2\text{NiZnFe}_{12}\text{O}_{22}$ –graphite–paraffin composites containing 80 wt% total filler. Graphite contents of 0, 8, 15, and 25 wt% are relative to the total ferrite–graphite filler.

The frequency dependence of the real and imaginary parts of the complex permittivity of the composites with different graphite contents is presented in Fig. 13. As the graphite content increases, the real part of the permittivity, ϵ' , increases considerably, from approximately 4.3 for the graphite-free composite to average values of 9, 35 and 65 for the composites containing 8, 15 and 25 wt% graphite, respectively. This increase can be attributed to the higher electrical conductivity, the gradual formation of conductive pathways, and interfacial polarization at the graphite–ferrite and particle–paraffin interfaces.

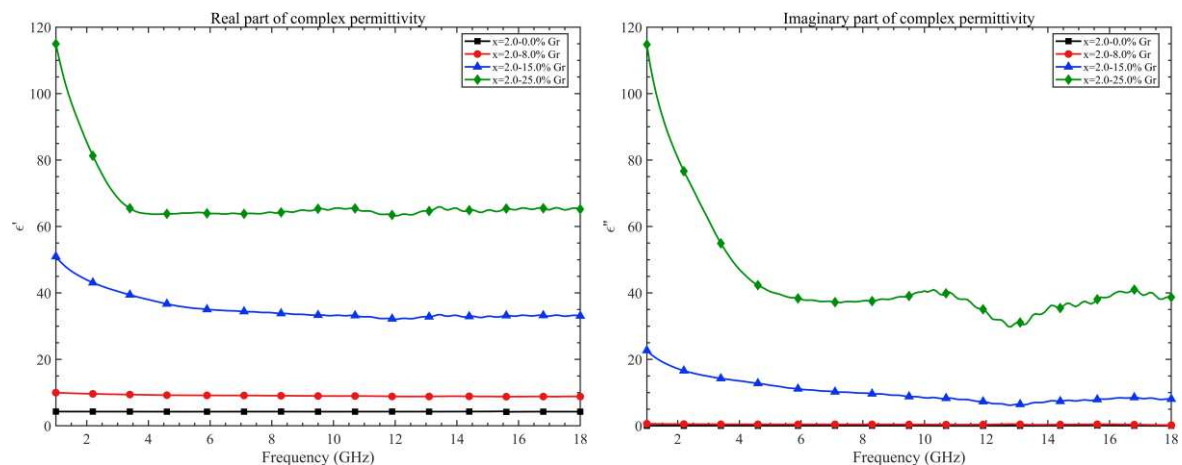


Fig. 13. the real part of permittivity ϵ' (left) and the imaginary part of permittivity ϵ'' (right) for ferrite-graphite-paraffin composites.

The simultaneous increase in ϵ'' indicates enhanced electromagnetic energy dissipation through conduction and polarization losses. The dielectric-loss capability should be evaluated using the dielectric loss tangent, $\tan \delta_\epsilon = \epsilon''/\epsilon'$. Fig. 14 shows the frequency dependence of the dielectric loss tangent over the frequency range of 1–18 GHz. In general, $\tan \delta_\epsilon$ increases with increasing graphite content, indicating an enhanced ability of the composites to dissipate electromagnetic energy. This improvement can mainly be attributed to the increase in conduction loss and the gradual formation of conductive pathways. Interfacial polarization at the graphite–ferrite and particle–paraffin interfaces may also contribute to this behavior. However, a higher dielectric loss tangent does not necessarily result in optimized microwave absorption, since an excessive graphite content may

degrade impedance matching and increase reflection at the absorber surface. Therefore, the graphite content should be optimized to achieve a suitable balance between impedance matching and electromagnetic-wave attenuation.

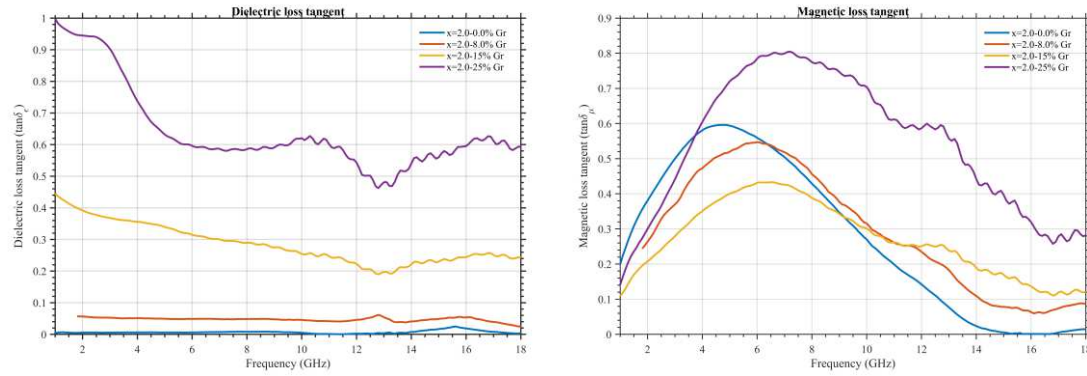


Fig. 14. Frequency dependence of the dielectric loss tangent (left) and the magnetic loss tangent (right) of the ferrite-graphite-paraffin composites.

The contour maps of the reflection loss of the ferrite-graphite-paraffin composites are compared in Fig. 15. The composite containing 8 wt% graphite shows a significant improvement in microwave absorption, with several frequency-thickness regions reaching $RL \leq -10$ dB and local minima below -20 dB. This improved performance can be attributed to a suitable balance between the complex permeability and permittivity. At this graphite content, the modified dielectric properties enable favorable impedance matching at different absorber thicknesses, resulting in broad frequency regions of strong microwave absorption, as shown in Fig. 15. In contrast, increasing the graphite content to 15 and 25 wt% considerably reduces the absorption performance, despite the relatively high dielectric loss tangents of these composites. Excessive graphite significantly increases the complex permittivity and promotes the formation of conductive pathways, while simultaneously decreasing the relative ferrite content and its magnetic contribution. This imbalance between the electric permittivity and magnetic permeability weakens impedance matching and causes a larger fraction of the incident wave to be reflected from the absorber surface. Consequently, the increased dielectric loss of these composites cannot be effectively utilized, because only a limited portion of the incident electromagnetic energy enters the absorber layer. Fig. 11 shows the effect of adding 8 wt% graphite to the $x = 2.0$ ferrite-based composite on the attenuation constant, α , and the real and imaginary parts of the normalized characteristic impedance, η_m/Z_0 . The addition of graphite decreases the real part of the normalized characteristic impedance. Therefore, the improved absorption performance cannot be directly attributed to improved intrinsic impedance matching. In contrast, the attenuation constant increases considerably over most of the measured frequency range, indicating an enhanced ability of the composite to dissipate electromagnetic energy, mainly through increased conduction loss and interfacial polarization. Accordingly, the improved microwave absorption of the composite containing 8 wt% graphite is mainly attributed to stronger internal attenuation and a more favorable balance between wave penetration and energy dissipation within the absorber layer. However, for a single-layer absorber backed by a metal plate, the overall impedance-matching condition should ultimately be evaluated using the normalized input impedance, Z_{in}/Z_0 , while considering its dependence on both frequency and absorber thickness.

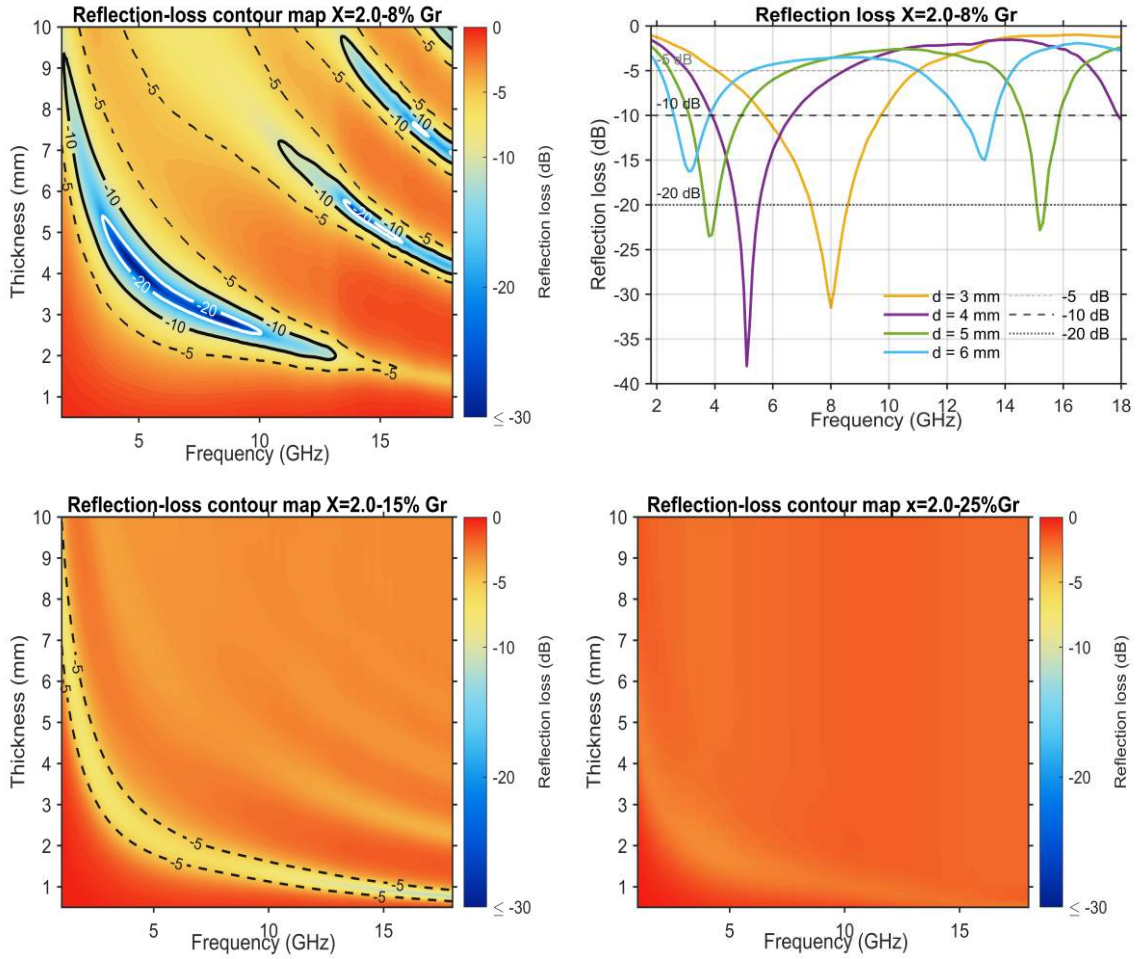


Fig. 15. Reflection-loss (RL) maps of the ferrite-graphite-paraffin composite samples with different mass percentages of graphite as functions of frequency and absorber thickness over the 1–18 GHz frequency range. The upper-right panel shows the RL spectra of the $x = 2.0$ -8% Gr composite at selected thicknesses.

Table 5 summarizes the microwave absorption properties of the composite samples containing 80 wt% ferrite–graphite filler dispersed in a paraffin matrix at different graphite contents. As summarized in Table 5, adding 8 wt% graphite increases the EAB from 3.7 to 4.0 GHz and reduces the corresponding absorber thickness from 4.5 to 3.0 mm. This simultaneous increase in absorption bandwidth and reduction in absorber thickness demonstrates improved absorption performance and highlights the beneficial role of graphite incorporation in achieving broadband microwave absorption with reduced material thickness.

Table 5. Microwave absorption characteristics of ferrite–graphite–paraffin composites based on the $x = 2.0$ ferrite with different graphite contents

Graphite content (wt%)	EAB (GHz) $RL \leq -10$ dB	d_{EAB} (mm)	f_{RLmin} (GHz)	RL_{min} (dB)	d_{RLmin} (mm)
0	3.7	4.5	11.1	-25	10
8	4	3	5.1	-38	4
15	0	-	13.8	-9	1
25	0	-	4.0	-3	2

To elucidate the origin of the absorption peaks and examine the relationship between absorber thickness and the frequency corresponding to the minimum reflection loss, the results were analyzed using the quarter-wavelength

cancellation model. According to this model, the matching thickness of a single-layer, metal-backed absorber is expressed as [25]

$$d_{\lambda/4} = \frac{(2n+1)c}{4f_{\lambda/4}\sqrt{|\varepsilon_r\mu_r|}}, \quad n = 0, 1, 2, \dots \quad (8)$$

where $d_{\lambda/4}$ is the matching thickness, $f_{\lambda/4}$ is the frequency at which the minimum reflection loss occurs, c is the speed of light in vacuum, and ε_r and μ_r are the complex relative permittivity and permeability evaluated at $f_{\lambda/4}$, respectively. Under this condition, the wave reflected from the absorber–metal interface travels a round-trip distance through the absorber and returns to the air–absorber interface with a phase difference of approximately π . Consequently, it interferes destructively with the wave reflected from the front surface, thereby reducing the overall reflection. The agreement between the thickness calculated using Equation (8) and that obtained from transmission-line theory provides further evidence supporting this mechanism; however, strong absorption also requires favorable impedance matching and sufficient dielectric and magnetic attenuation within the absorber.

Fig. 16 compares the frequencies corresponding to the minimum reflection loss, f_{RLmin} , with the matching frequencies predicted from the quarter-wavelength condition, $f_{\lambda/4}$, for the $x = 2.0$ composites containing 0 and 8 wt% graphite at absorber thicknesses of 3 and 5 mm. For the composite containing 8 wt% graphite, good agreement is observed between the f_{RLmin} and predicted matching frequencies. At $d = 3$ mm, the minimum reflection loss occurs at approximately 8.0 GHz, which is close to the predicted matching frequency of about 8.2 GHz. At $d = 5$ mm, two reflection-loss minima appear near 3.8 and 15.2 GHz. These minima are consistent with the matching frequencies obtained from different odd-order quarter-wavelength conditions, corresponding to different values of n . Acceptable agreement is also observed for the graphite-free composite at $d = 5$ mm.

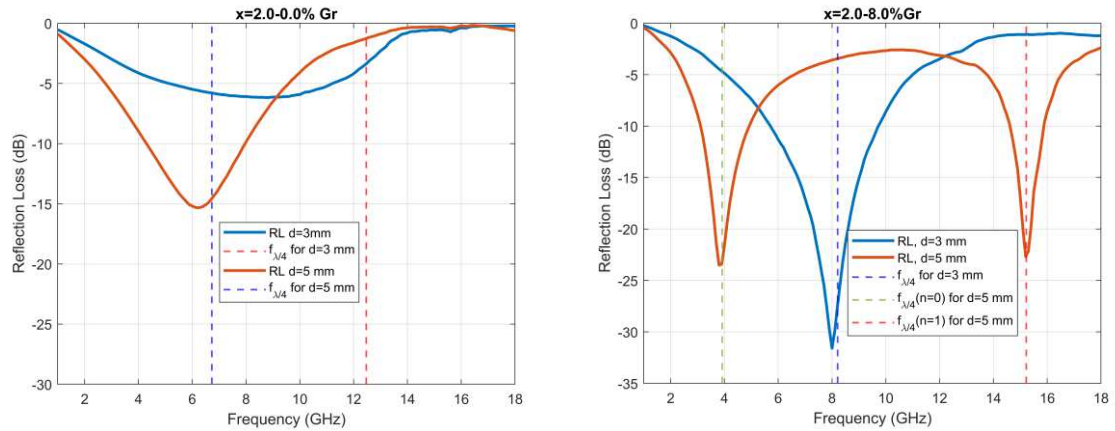


Fig. 16. Comparison of the reflection-loss spectra and the quarter-wavelength matching frequencies, ($f_{\lambda/4}$), for the $x = 2.0$ composites containing 0 and 8 wt% graphite at absorber thicknesses of 3 and 5 mm.

The vertical dashed lines indicate the matching frequencies calculated using Eq. (8).

However, for the graphite-free composite at $d = 3$ mm, the calculated matching frequency of approximately 12.5 GHz does not coincide with any distinct minimum in the reflection-loss curve. Instead, only a broad and relatively shallow absorption region appears at lower frequencies. This difference shows that satisfying the quarter-wavelength phase condition alone does not necessarily produce strong microwave absorption. Favorable impedance matching and sufficient attenuation of the wave inside the absorber are also required. As shown in Fig. 11, the relatively low attenuation constant, α , near 12.5 GHz limits electromagnetic energy dissipation and prevents the formation of a pronounced reflection-loss minimum.

The remaining small differences between $f_{\lambda/4}$ and f_{RLmin} can be attributed to the dispersive and lossy nature of ε_r and μ_r , as well as to the approximate form of Eq. (8). In this equation, $\sqrt{|\varepsilon_r\mu_r|}$ is used as an effective term for

the refractive index; therefore, this approximation cannot fully reproduce the actual phase constant, β . Overall, the observed agreement confirms destructive interference under the quarter-wavelength condition, while the depth of the reflection-loss minima is simultaneously governed by phase interference, impedance matching, and dielectric and magnetic attenuation.

4. Conclusion

Simultaneous Ni–Zn substitution changed the magnetic resonance behavior of the Co₂Y-based hexaferrites. The coercive field decreased with increasing substitution, but the resonance frequency changed nonmonotonically. It also did not follow the trend of the magnetization. The distributed Kittel–Gilbert model successfully described the broad and asymmetric permeability spectra. This behavior is consistent with a distribution of local resonance conditions. The $x = 2$ sample required a higher calcination temperature, which also affected its microstructure. Therefore, its behavior cannot be attributed to composition alone.

The graphite-free $x = 2$ composites showed an EAB of 3.7 GHz at a thickness of 4.5 mm. Adding 8 wt% graphite relative to the filler mass increased the EAB to 4.0 GHz and reduced the required thickness to 3.0 mm. A minimum RL of –38 dB was obtained at 5.1 GHz and 4.0 mm. Further increasing the graphite content to 15 or 25 wt% reduced the absorption performance, even though the dielectric loss increased. This shows that higher dielectric loss alone does not guarantee better absorption. The thickness-dependent RL minima were also consistent with approximate quarter-wavelength interference. However, this condition alone was not sufficient to produce strong absorption.

Overall, Ni–Zn substitution provides a way to tune the magnetic resonance, while a controlled amount of graphite can improve absorption at lower thicknesses. The combination of these two approaches can therefore be useful for adjusting the electromagnetic response of Y-type hexaferrite absorbers.

Acknowledgement

The financial support of this work by the Research Council of Malek Ashtar University of Technology is gratefully acknowledged.

Data availability

Data will be made available on request.

References

- [1] Mohammed, Ibrahim, et al. "Review on Y-type hexaferrite: synthesis, characterization and properties." *Applied Surface Science Advances* 16 (2023): 100416. doi.org/10.1016/j.apsadv.2023.100416.
- [2] Khanduri, H., et al. "Structural, dielectric, magnetic, and nuclear magnetic resonance studies of multiferroic Y-type hexaferrites." *Journal of Applied Physics* 112.7 (2012). doi: 10.1063/1.4754532.
- [3] Popov, Maksym, et al. "Nonlinear magnetoelectric effects in Y-type hexaferrite microwave resonators." *Journal of Applied Physics* 128.11 (2020). doi: 10.1063/5.0021593.
- [4] Suthar, Mukesh, et al. "Structural, magnetic, and biocompatibility evaluations of chromium substituted barium hexaferrite (Co₂–Y) for hyperthermia application." *Materials Chemistry and Physics* 296 (2023): 127348. doi: https://doi.org/10.1016/j.matchemphys.2023.127348.
- [5] Kocsis, Vilmos, et al. "Magnetization-polarization cross-control near room temperature in hexaferrite single crystals." *Nature Communications* 10.1 (2019): 1247. doi: 10.1038/s41467-019-09205-x.
- [6] Popov, Maksym, et al. "Y-type hexagonal ferrite-based band-pass filter with dual magnetic and electric

field tunability." *Scientific Reports* 13.1 (2023): 1179.

doi: 10.1038/s41598-023-28279-8.

[7] Pullar, Robert C. "Hexagonal ferrites: a review of the synthesis, properties and applications of hexaferrite ceramics." *Progress in Materials Science* 57.7 (2012): 1191-1334. doi: 10.1016/j.pmatsci.2012.04.001.

[8] Adeela, N., et al. "Structural and magnetic response of Mn substituted Co₂Y-type barium hexaferrites." *Journal of Alloys and Compounds* 686 (2016): 1017-1024.

doi: <https://doi.org/10.1016/j.jallcom.2016.06.239>.

[9] Samadpour, Elham, Esmail Kiani, and Mohammad Hosein Shams. "Microwave permeability and electromagnetic wave absorption properties of Co₂Y nanocomposites." *Materials Science and Engineering: B* 298 (2023): 116825.

doi: 10.1016/j.mseb.2023.116825.

[10] Farzin, Y. Alizad, O. Mirzaee, and A. Ghasemi. "Influence of Mg and Ni substitution on structural, microstructural and magnetic properties of Sr₂Co_{2-x}Mg_{x/2}Ni_{x/2}Fe₁₂O₂₂ (Co₂Y) hexaferrite." *Journal of magnetism and magnetic materials* 371 (2014): 14-19.

doi: 10.1016/j.jmmm.2014.07.007.

[11] JMohammed, J., et al. "Crystal structure refinement and the magnetic and electro-optical properties of Er³⁺-Mn²⁺-substituted Y-type barium hexaferrites." *Ceramics International* 47.13 (2021): 18455-18465.

doi: <https://doi.org/10.1016/j.ceramint.2021.03.169>.

[12] Arshad, Muhammad, et al. "Investigation of crystal structure, electrical and dielectric response of Ga³⁺ substituted Sr-Ni Y-type hexaferrites as a suitable material for high frequency applications." *Ceramics International* 49.12 (2023): 21036-21049.

doi: <https://doi.org/10.1016/j.ceramint.2023.03.239>.

[13] Mohammed, Ibrahim, J. Mohammed, and A. K. Srivastava. "Effect of Dy³⁺-Cu²⁺ doping on structural, magnetic and electromagnetic properties of Co₂Y-type hexaferrite." *Ceramics International* 49.10 (2023): 16467-16476.

doi: <https://doi.org/10.1016/j.ceramint.2023.02.008>.

[14] Kanwal, Shumaila, et al. "Study of structural, spectral and dielectric properties of Dy substituted Sr₂Co₂Fe_{12-x}Dy_xO₂₂ Y-type hexaferrites via sol-gel auto-combustion method for microwave absorption application." *Materials Chemistry and Physics* 322 (2024): 129558.

doi: <https://doi.org/10.1016/j.matchemphys.2024.129558>.

[15] Jafari, A., et al. "The role of Sn and Zn substitutions on the magnetic properties of nanosized Y-type hexaferrite prepared by sol-gel auto-combustion method." *Physica Scripta* 99.3 (2024): 035933.

doi: 10.1088/1402-4896/acfc89.

[16] Won, Mi Hee, and Chul Sung Kim. "Magnetic properties of Ni substituted Y-type barium ferrite." *Journal of Applied Physics* 115.17 (2014).

doi: 10.1063/1.4860939.

[17] Mahmood, S. H., et al. "Structural properties and hyperfine interactions in Co-Zn Y-type hexaferrites prepared by sol-gel method." *Ceramics International* 40.4 (2014): 5231-5236.

doi: 10.1016/j.ceramint.2013.10.092.

[18] Malik, Huma, et al. "Structural, spectral, thermal and dielectric properties of Nd-Ni co-doped Sr-Ba-Cu hexagonal ferrites synthesized via sol-gel auto-combustion route." *Ceramics International* 44.1 (2018): 605-612.

doi: 10.1016/j.ceramint.2017.09.219.

[19] Roberts, A. P., Yulong Cui, and K. L. Verosub. "Wasp-waisted hysteresis loops: mineral magnetic characteristics and discrimination of components in mixed magnetic systems." *Oceanographic Literature Review* 4.43 (1996): 384.

doi: 10.1029/95JB00672

[20] Neupane, D., et al. "Synthesis and magnetic study of magnetically hard-soft SrFe_{12-y}Al_yO_{19-x} Wt.% Ni_{0.5}Zn_{0.5}Fe₂O₄ nanocomposites." *AIP Advances* 7.5 (2017).

doi: 10.1063/1.4978398

[21] Kim, Chin Mo, et al. "The effect of precursor types on the magnetic properties of Y-type hexa-ferrite composite." *Journal of Applied Physics* 117.17 (2015).

<https://doi.org/10.1063/1.4918690>

[22] Fiorillo, F., and C. Beatrice. "A comprehensive approach to broadband characterization of soft ferrites." *International Journal of Applied Electromagnetics and Mechanics* 48.2-3 (2015): 283-294.

doi: 10.3233/JAE-152000.

[23] Kiani, Esmail, Amir SH Rozatian, and Mohammad H. Yousefi. "The Effects of Doping on Crystal Structure, Magnetic and Microwave Properties of SrFe_{12-2x}La_x(Mn_{0.5}Zr_{0.5})_xO₁₉ Nanoparticles." *Journal of superconductivity and novel magnetism* 26.3 (2013): 733-738.

<https://doi.org/10.1007/s10948-012-1809-5>

[24] Zhao, Rui, et al. "Recent progress of carbon-based magnetic fibers for electromagnetic wave absorption." *Carbon* 229 (2024): 119513.

doi: <https://doi.org/10.1016/j.carbon.2024.119513>.

[25] Suetake, K. "Application of ferrite to electromagnetic wave absorber and its characteristics." *IEEE Transactions on Microwave Theory and Techniques* 19.1 (1971): 65-72.

doi: 10.1109/TMTT.1971.1127446.