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Research articles

Magnetically tunable composite ferrite-dielectric microwave elements

Anatolii Belous[a], Oleksandr Fedorchuk[a], Sergii Solopan[a,*], Maksym Popov[b], Igor Zavislyak[b]

[a] Solid State Chemistry Department V.I. Vernadskii Institute of General and Inorganic Chemistry of the NAS of Ukraine Kyiv, Ukraine
[b] Faculty of Radiophysics,Electronics and Computer Systems Taras Shevchenko National University of Kyiv Kyiv, Ukraine

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ABSTRACT

Multiphase microwave dielectrics with high electrical quality and adjustable thermal stability of electrical properties have been investigated on the basis of barium polytitanates. The nature of high Q-factor and the opportunity to manage the thermal stability of multiphase systems is explained. Composite resonance elements based on high-quality multiphase dielectric materials and ferromagnetic film have been developed. It is shown that composite resonators are characterized by high Q-factor and the ability to control the resonant frequency by a magnetic field, which allows them to be used in various tunable microwave devices.

1. Introduction

Nowadays the single-phase dielectrics with low dielectric losses ($tg\delta$) or high dielectric quality ($Q = 1/tg\delta$) in the microwave range and with high thermal stability of physical properties have become one of the main materials for the creation of microwave devices and components [1–5]. For example they are used for radiolocation, wireless and satellite communications. The use of dielectrics on the base of single-phase material is caused by the fact that the presence of additional phases almost always causes the increasing of dielectric losses.

The electrophysical properties of such dielectrics are not sensitive to external electric and magnetic fields. And the main trends and requirements to the new microwave components are miniaturization, increase in versatility, performance, smart components development and the ability to change own parameters of devices under the influence of external fields. Nevertheless, the dielectrics with low dielectric losses and with properties that are insensitive to the external electric and magnetic fields remain the basis for the creation of a number of microwave technics components (filters, diplexers, solid-state generators).

There are works [6–9] where authors are trying to combine low dielectric losses with the possibility to manage properties of materials by external fields. It is suggested to reach the compromise between the low dielectric losses and properties management by a combining the abovementioned dielectrics and materials that change their properties due to the influence of temperature, other physical influences or by the development of elements with the use of microelectromechanical systems (MEMS), etc. It should be remembered that the setting with electric and magnetic fields is the least inertial among them. Therefore, the highest priority must be given to the field control. Among the microwave materials that are sensitive to external electromagnetic influence ferrites are distinguished.

Ferrites are materials based on oxide systems, magnetics with orderly located magnetic moments in their structure. It is known that these materials change their high frequency magnetic susceptibility under the influence of magnetic fields. Also in ferrites the ferromagnetic resonance (FMR) phenomenon can be observed under certain conditions and this phenomenon is accompanied by the resonant absorption of the energy of an electromagnetic field interacting with ferrite. Compared with ferromagnetics (an another class of magnetics with strong magnetic properties) ferrites have weaker magnetic properties but due to specific features of their structure they are electrically low-conductive materials (ferromagnetics almost always are conductors or high-conductive semiconductors). This fact allows to decrease the eddy current losses and it is one of the main reasons why ferrites have become widespread in the microwave engineering as the components for the high-frequency devices: isolators, circulators, phase shifters, various magnetooptical devices etc. However, microwave losses in ferrites still far exceed losses in non-magnetic dielectrics. Therefore composite elements that combine high-Q, field-insensitive microwave dielectrics with ferrites are of considerable interest. In presented work it is suggested to use the ferrite component in a form of a film deposited on the surface of a cylindrical dielectric resonator (DR) made of a non-magnetic dielectric, that far exceed a magnetic film by volume.

It should be noted that we have previously developed composite elements in the form of resonators based on α-Al_2O_3 that is characterized by the low value of the dielectric constant $\varepsilon \sim 10$ and nickel ferrite $NiFe_2O_4$ [10]. It was shown that obtained composite elements are capable of changing their properties – a feature necessary for practical

* Corresponding author.
E-mail address: solopan@ukr.net (S. Solopan).

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applications.

One more important objective of microwave components, apart from the opportunity of adjustment by magnetic fields, is the providing of thermal stability of devices. It is known that the dielectric constant temperature factor (τ_ε) in the single-phase materials is a constant value and often it doesn't allow to compensate for a thermal instability of microwave circuits. For practical applications usually materials with adjustable values of τ_ε are needed. All this factors limit the practical use area of DR based on single-phase microwave dielectrics. Therefore it is important to search the ways to create microwave materials with the precise control of thermal stability by changing the chemical composition and also the ways of development of resonant elements with the frequency tunable by external electromagnetic field.

It is known that materials based on $BaTi_4O_9$ are characterized by the relatively high value of dielectric constant ($\varepsilon = 36$), Q-factor ($Qf \approx$ 35000–40,000 GHz) and thermal stability (the dielectric constant temperature factor $\tau_\varepsilon = -(3\text{–}4)\cdot 10^{-5}$ K^{-1}) [1]. Authors [11,12] showed that the additives of a number of metal oxides, in particular of zinc oxide, allows to impact on both the thermal stability of $BaTi_4O_9$ electrophysical properties and Q-factor value (which appears to increase). But the nature of this effect was not clear at that time.

In this work a material based on $BaTi_4O_9$-ZnO multiphase system was used as the non-magnetic component and solid solutions of nickel zinc ferrite with the spinel structure $Ni_{1-x}Zn_xFe_2O_4$ (magnetically soft materials with high saturation magnetization, effective magnetic permeability and coercive force [13]), were used as the magnetic component.

The aim of this investigation is a study of creation regularities of high-Q multiphase microwave dielectrics based on barium tetratitanate and zinc oxide, finding out possibilities of influence on the thermal stability of electrophysical properties and also of development of composite resonant microwave elements based on multiphase dielectric materials with high Q-factor and ferrimagnetic film made from nickel-zinc ferrite that are capable of controlling a resonance frequency and energy absorption of the composite by external magnetic field.

2. Experimental details

2.1. Synthesis of barium tetratitanate $BaTi_4O_9$ – ZnO

Barium tetratitanate was manufactured by the method of solid-state synthesis. As initial reagents were used $BaCO_3$, TiO_2 and ZnO of *puriss. spec.* qualification. On the first stage a fine powder of $BaTi_4O_9$ was synthesized. On the second stage, ZnO was being added to the powder in different amount at the temperature of 900 °C for 3 h. The sintering of ceramics samples was being carried out at the temperature of 1320–1340 °C for 2 h.

2.2. Synthesis of nanoscaled particles of nickel-zinc ferrites $Ni_{1-x}Zn_xFe_2O_4$ solid solutions with spinel structure

Synthesis of nanoscaled particles of nickel-zinc ferrites solid solutions was carried out by the method of coprecipitation from aqueous solutions of hydroxides [10]. An aqueous solutions of nitrates $Fe(NO_3)_3$, $Ni(NO_3)_2$ and $Zn(NO_3)_3$ were used as initial reagents and 1 M solution of NaOH was used as precipitator. The precipitation of salts was carried out successively at different values of pH selected individually for each metal. At the first stage a precipitation of salt $Fe(NO_3)_3$ was carried out at pH value of 4 ÷ 4.5. Then a precipitation of salt $Zn(NO_3)_3$ at pH = 7.0 ÷ 7.2 was carried out with continuous stirring. After the precipitation of $Fe(NO_3)_3$ and $Zn(NO_3)_3$ pH of mother solution was increased to 8.5 ÷ 8.7 and the solution of salt $Ni(NO_3)2$ was precipitated successively with continuous stirring.

After the precipitation of all the components the aging of precipitates was carried out. For this the suspension was being boiled for 1 h. During the boiling an initial volume of the suspension was maintained constant.

The obtained precipitate was filtered and washed by distilled water. The product was dried in a drying cabinet at the temperature 110–120 °C. The final product was obtained after the calcination of precipitates in a muffle furnace in the atmosphere of air at a wide temperature range 500–800 °C.

The size and morphology of ferrites particles were determined by transmission electron microscopy on a *JEM* 1400 *Jeol* instrument.

Synthesized powders of $Ni_{1-x}Zn_xFe_2O_4$, x = 0, 0.25, 0.5, 0.75, 0.8, 0.85, 0.9, 0.95, 1, were investigated by the X-ray analysis on a "ДРОН-4" diffractometer (CuKα radiation). The degree of crystallinity of synthesized nanoscaled ferrites particles was determined by a methodic presented in [14].

Measurements of magnetic hysteresis loops of obtained powders were made using a vibrating magnetometer *LDJ*-9500.

2.3. Deposition of nickel-zinc ferrites films $Ni_{0.5}Zn_{0.5}Fe_2O_4$ with the spinel structure on non-magnetic dielectric resonator

Ferrite films were deposited on cylindrical dielectric resonators using suspensions consisted of nanoscaled nickel-zinc ferrites powders and organic constituent. An organic constituent was formed by the photopolymer (industrial UV glue Permabond UV630, with the curing wavelength 365–410 nm). The suspensions were mixed using a homogenizer *IKA T*10 *Standard* and deposited to the end face of resonators made of barium tetratitanate in the way described in [10]. Ferrites films were being polymerized under the UV light (Fig. 1) either without the magnetic field or with the magnetic fields in 2 different directions: parallel and normal to resonators axis (one of three variants for any given film). The strength of bias field was H = 2900 E.

2.4. Investigation of electrophysical properties properties of composite resonant elements

Energy transmission spectra of manufactured composite resonators were measured by Agilent N5230A PNA vector analyzer. Properties of composite resonators were investigated in the centimeter wavelength range (X-band) in the traveling wave mode. A measuring cell was an X-band waveguide with the cross-sectional area of 23·10 mm^2. Resonators were placed on the wide wall of the waveguide, their optimal position inside the waveguide was selected manually. A glass substrate separates the resonators from the metal walls of waveguide and reduced the eddy currents losses. Investigation was conducted similar to the one in [10].

3. Results and discussions

On the Fig. 2 the X-ray pattern of synthesized $BaTi_4O_9$ and of the multiphase sample based on $BaTi_4O_9$ – ZnO are presented. When ZnO is entered an additional phase appears. The additional phase that appears in the system $BaTi_4O_9$ – ZnO was initially decrypted as $Ba_3Zn_7Ti_{12}O_{34}$

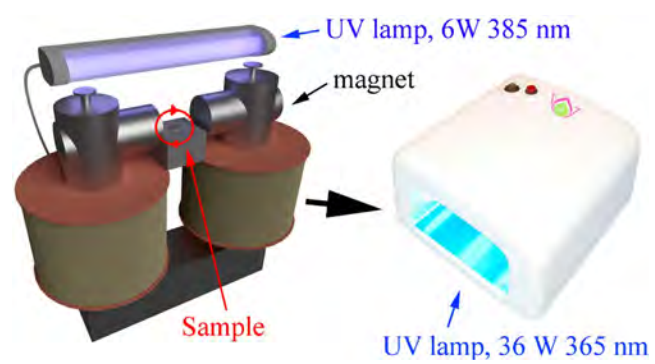

Fig. 1. Polymerization of a magnetic film in magnetic field under UV lamps.

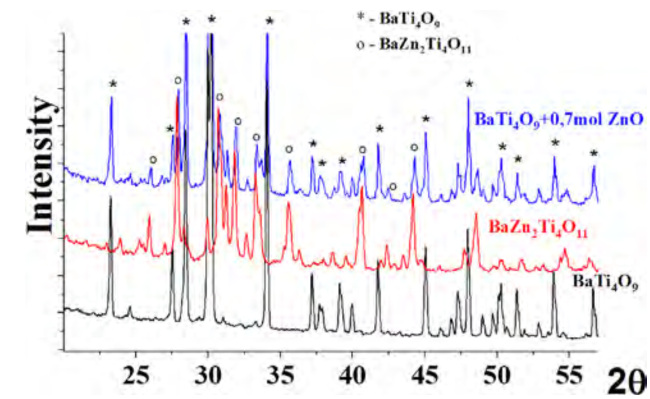

Fig. 2. XRD patterns of ceramics samples of $BaTi_4O_9$, $BaTi_4O_9$ – ZnO, and ceramics with the $BaZn_2Ti_4O_{11}$ additional phase.

($BaZn_{21/3}Ti_4O_{111/3}$) [15]. Later the investigation of a phase diagram of the BaO-ZnO-TiO_2 system conducted by authors in [16] has shown that the additional phase in the $BaTi_4O_9$ – ZnO system possibly corresponds to the $BaZn_2Ti_4O_{11}$ phase. Therefore $BaZn_2Ti_4O_{11}$ based materials were also synthesized by the solid-state reactions method, and the X-ray patterns of obtained materials are presented on the Fig. 2 alongside with $BaTi_4O_9$ – ZnO.

It can be observed from the Fig. 2 that peaks of additional phase occurring in the system $BaTi_4O_9$ – ZnO and peaks of the $BaZn_2Ti_4O_{11}$ phase coincide. Hence, the interaction of barium tetratitanate and zinc oxide can be described by the following reaction:

$$BaTi_4O_9 + 2xZnO = (1 - x)BaTi_4O_9 + xBaZn_2Ti_4O_{11}.$$

Regardless of amount of zinc oxide entered in barium tetratitanate the $BaTi_4O_9$–ZnO system contains the $BaTi_4O_9$ and $BaZn_2Ti_4O_{11}$ phases only. A number of interesting regularities appear when ZnO is entered in $BaTi_4O_9$ should be noted: a) if the temperature coefficient of dielectric constant at the microwave range is within the values of $-(3–4)\cdot10^{-5}\ K^{-1}$ the ZnO entering can change τ_ε from negative to positive values; b) when the additional $BaZn_2Ti_4O_{11}$ phase occurs the Q-factor rises which is unexpected because usually the appearance of an additional phase in a microwave dielectric leads to additional losses (Q-factor decreases).

To find out the nature of the influence of zinc oxide on the properties of $BaTi_4O_9$ the properties of $BaZn_2Ti_4O_{11}$ were investigated in the microwave range. It was shown that $BaZn_2Ti_4O_{11}$ is characterized by $\varepsilon = 20$, $\tau_\varepsilon = +2\cdot10^{-5}\ K^{-1}$ and high Q-factor Q ≥ 9000 at the frequency of 10 GHz. These data indicate that when zinc oxide is entered in barium tetratitanate the new phase ($BaZn_2Ti_4O_{11}$) with a higher (than of $BaTi_4O_9$) Q-factor and positive τ_ε appears. This phenomenon provides an increase of Q-factor of two-phase materials and also an increase of the properties thermal stability due to effect of volumetric thermal compensation. In addition a τ_ε value can be changed from negative to positive values by varying of a concentration of ZnO in the $BaTi_4O_9$–ZnO system.

On the base of the $BaTi_4O_9$ – ZnO system materials the high-Q dielectric microwave resonators may be were developed that can be used in a design of various devices (filters, solid-state generators, etc.).

But such dielectric resonators unable to change their resonance frequency under the influence of external electric or magnetic fields what is necessary for the development of heterodyne receivers, for the increase of versatility and compactness of radiolocation systems and other communications.

The theoretical calculations of an interaction of an electromagnetic field and the composite resonator were made to find out the possibilities of the creation of composite dielectric resonators that are capable to change resonance frequency due to the influence of magnetic field and consists of high-Q dielectric resonator based on the multiphase $BaTi_4O_9$–ZnO system with the magnetic film deposited on it.

The influence of a ferrite film on properties of a composite DR may be found from the perturbations theory applied to the Maxwell's equations. From the Maxwell's equations in the integral form after certain mathematical transformations and integration by volumes of a dielectric and inserted perturbation the following expression can be obtained [17]

$$\int_V \nabla\bullet(H_2 \times E_1^* + H_1^* \times E_2)dV = j\Big[(\omega_2 - \omega_1) \bullet \int_V (\hat{\varepsilon}_1 E_2 \bullet E_1^* + \hat{\mu}_1 H_2 \bullet H_1^*)dV + \omega_2 \int_V (\Delta\varepsilon E_2 \bullet E_1^* + \Delta\mu H_2 \bullet H_1^*)dV\Big]$$

where $\Delta\varepsilon = \hat{\varepsilon}_2 - \hat{\varepsilon}_1$, $\Delta\mu = \hat{\mu}_2 - \hat{\mu}_1$. The lower index corresponds to the unperturbed and perturbed states of a system respectively

Further it can be proved that a change of the complex resonator frequency due to a perturbation is given by the formula

$$\frac{\omega_2 - \omega_1}{\omega_1} \approx -\frac{\int_{V_s} (\Delta\varepsilon E_1^*\cdot E_2 + \Delta\mu H_1^*\cdot H_2)dV}{2\int_{V_c} \Delta\varepsilon E_1^*\cdot E_2 dV} \tag{2}$$

where $\Delta\varepsilon$, $\Delta\mu$ is a difference between relevant parameters of a DR and ferrite, V_s is a volume of a ferrite, V_c is a volume of a non-magnetic substituent. Using known formulas for the field of the operating mode of a free dielectric resonator we can obtain an analytical expression describing the relative frequency shift [17].

In the work [18] it is shown that for the effective excitation of FMR in a composite resonator inside a waveguide the eigen modes $E_{\pm 11\delta}$ of dielectric are the most effective, analytical expressions for the resonance frequency shift under the influence of the field for the named dielectrics modes were obtained:

$$\frac{\Delta f_r}{f_{r0}} = -(\chi' \pm 0.837\chi'_a)\frac{V_S}{V_C} \tag{3}$$

where χ', χ_a' are real components of a tensor of the magnetic susceptibility [19]; Δf_r is a shift of a resonance frequency of a composite resonator; f_{r0} is a unperturbed resonance frequency of a composite resonator without the external biasing field. Different signs in (3) correspond to different directions of polarization of the unperturbed mode of a resonator, namely the sign "+" corresponds to the eigen mode $E_{+11\delta}$ with the right circular polarization and sign "−" corresponds to the mode $E_{-11\delta}$ with the left circular polarization.

The above ratios show that the relative shift of the resonance frequency (Δ_{fr}/f_{r0}) is directly proportional to the volume (thickness) of a magnetic film, its magnetic susceptibility magnetization and therefore to magnetization.

In the other hand a composite DR must be characterized by the relatively high Q-factor value. The calculations show that the change of composite structure's inverse Q-factor is proportional to the volume of a magnetic film [18]:

$$\Delta(1/2Q) = (\chi'' \pm 0.837\chi''_a)V_S K/V_C \tag{4}$$

where $K \approx 1$ is a dimensionless coefficient.

It is also important to find the optimal ratio between the dimensions of a high-Q DR and a ferrimagnetic film to provide both the high Q-factor of a composite resonator and the frequency shift under the influence of the magnetic field. In our case a method of the film applying on a dielectric resonator and the choice of the optimum thickness (near 100 µm) were determined in the work [10].

One more consequence of the ferrite influence on the properties of a composite resonator is the non-reciprocity of electrophysical properties with respect to the direction of the electromagnetic wave propagation through the structure [20].

According to the abovementioned criteria of magnetic material selection, nickel-zinc ferrites $Ni_{1-x}Zn_xFe_2O_4$ solid solutions were chosen as the magnetic phase due to their high magnetization, small coercive force and relatively low magnetic losses. Although zinc is a paramagnetic, its addition can lead to an increase in magnetization compared to nickel ferrite. Ni^{2+} ions in nickel ferrite occupy B sites in the spinel structure (octahedral positions) and material has a structure of normal spinel. At the same time Zn^{2+} ions has larger ion radius than nickel and prefer to occupy A sites [20] forming the inverse spinel structure. The $Ni_{1-x}Zn_xFe_2O_4$ solid solutions form the structure that has normal distribution of cations to Zn and inverted for Ni [21,22,23]

$$Zn_x \overleftrightarrow{Fe^{3+}_{1-x}} [\overrightarrow{Ni^{2+}_{1-x}} + \overrightarrow{Fe^{3+}_{1+x}}]O_4$$

Magnetic structure of the materials changes. For nickel ferrite the superexchange interaction A-O-B (A site ions – oxygen – B site ions) is dominant and the iron in the A site will be aligned in an antiparallel direction with respect to spins of cations on B sites [20]. Therefore the nickel ferrite is a ferrimagnet with total magnetic moment equal to the difference between magnetic moments in A- and B-sites. For zinc ferrite the superexchange interaction B-O-B is present only. Hence this material is antiferromagnet with zero net magnetic moment. But for $Ni_{1-x}Zn_xFe_2O_4$ structures values of named interactions can be changed via composition variations. The iron aligning can be changed and change magnetization of material dramatically. So, for the $Ni_{1-x}Zn_xFe_2O_4$ structure the resulting magnetic moment equals to the magnetic moments difference of octahedral and tetrahedral spinel sublattices too, but taking into account a variable direction of iron ions magnetic moments. According to literature, if one marks the magnetic moment of the Ni^{2+} ion or of an another magnetic divalent metal ion as m and the moment of the Fe^{3+} ion as $f = 5$ then the magnetization of one formula unit of ferrite at × range of 0–0.5 with the good precision equals [21] to

$$M = [(1 - x)m + (1 + x)f] - [0 + (1 - x)f] = 10x + m(1 - x) \quad (5)$$

For the ferrite with nickel the maximal experimental value of the unit cell magnetic moment approaches that of one iron ion i.e. 5 μ_B.

X-ray patterns of particles of the nickel-zinc ferrite with composition $Ni_{0,5}Zn_{0,5}Fe_2O_4$ synthesized by the precipitation from aqueous solutions after the thermal treatment at different temperatures are presented on the Fig. 3a. X-ray studies showed that single-phase particles are formed in one stage at the temperature of 500 °C and above. But increase of the temperature affects the crystallinity degree and dimensions of the product particles. The crystallinity degree of synthesized particles after

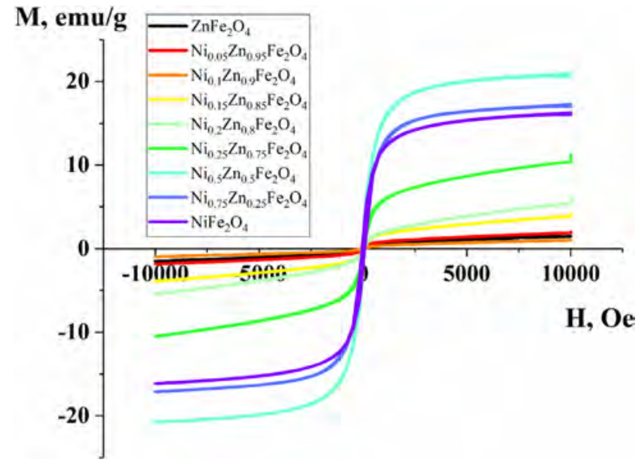

Fig. 4. Magnetic hysteresis loops of powders of nickel-zinc ferrites solid solutions.

the thermal treatment at different temperatures is 32.7% at 500 °C; 34.2% at 600 °C; 41.7% at 650 °C; 51.7% at 700 °C; 89.1% at 800 °C. We decided to use particles synthesized at the temperature of 800 °C. The crystallinity degree of ferrite particles after the thermal treatment at the temperature of 800 °C was 89%, the average size of particles according to results of micrographs was 50 nm.

The magnetic hysteresis loops of synthesized powders were measured (Fig. 4). As it can be seen the highest value of the saturation magnetization is observed for the $Ni_{0.5}Zn_{0.5}Fe_2O_4$ ferrite. A change of the specified ratio between the ions of nickel and zinc leads to the decrease of the saturation magnetization in the formed partially inverted spinel structure.

Composite resonant elements transmission spectra recorded at different values and directions of external magnetic field are presented on the Fig. 5. The Fig. 5a represents the maximal frequency shift and Fig. 5b represents the maximal isolation value for resonators, the dashed curves on the Fig. 5b show the reverse transmission (S_{12}) of resonators, magnetic field values are the same as for the “direct” transmission (S_{21}). The value of the frequency shift under the influence of the field of 0–3500 Oe is 71 MHz at the frequency around 11 GHz (Fig. 5a) for the case when the magnetic film is polymerized by the normal magnetic field. In the case of polymerization in the parallel magnetic field the frequency shift is 41 MHz.

Additional studies of composite resonant elements in the waveguide

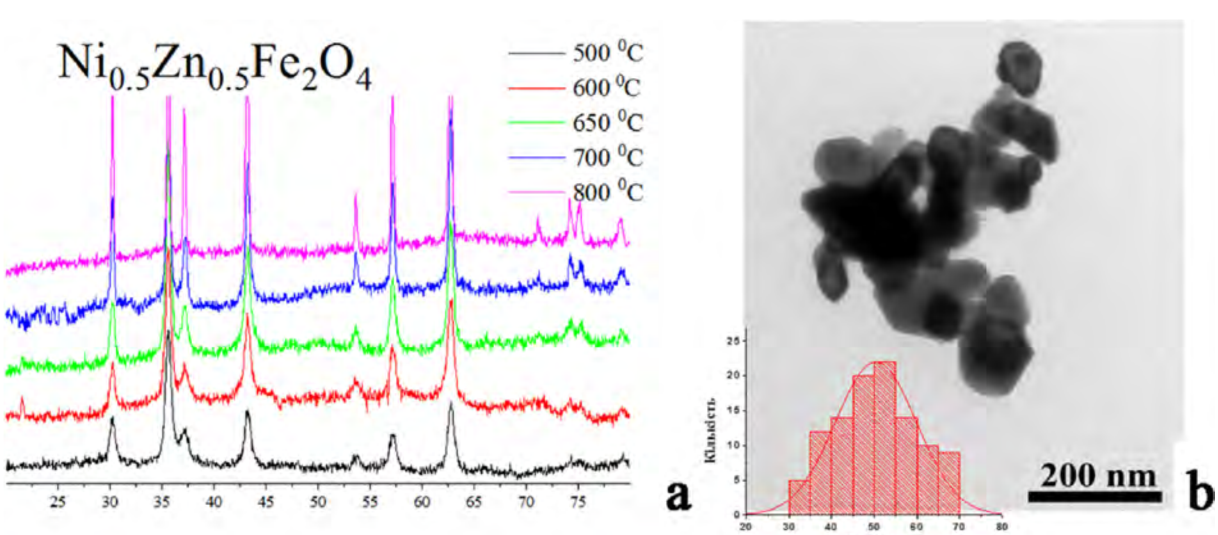

Fig. 3. XRD patterns and TEM images of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ particles.

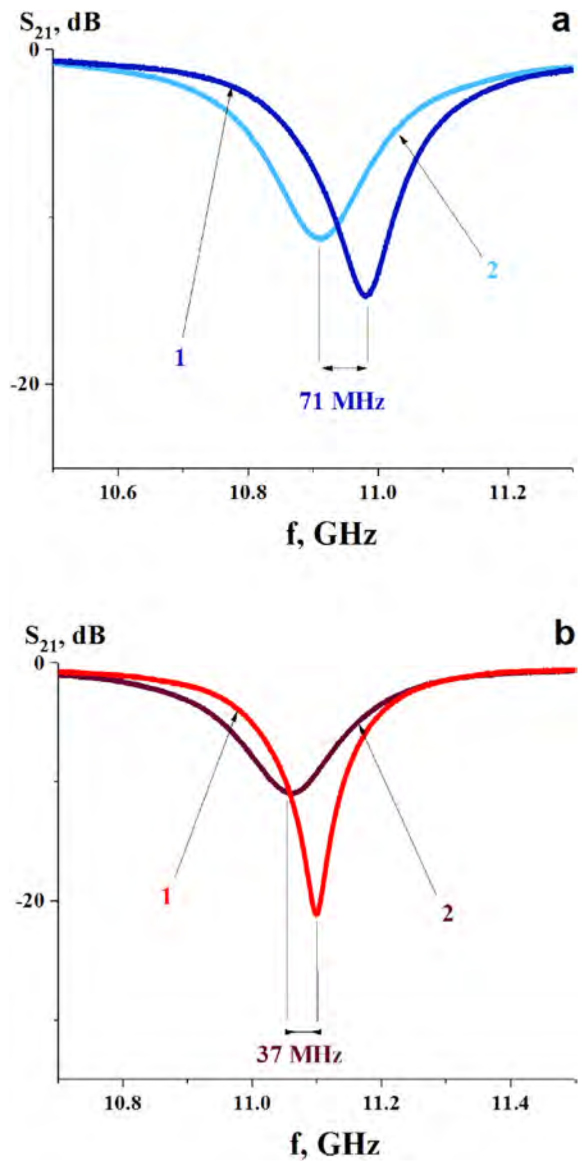

Fig. 5. Energy transmission spectra of the composite resonators with ferrite films polymerized in the magnetic field directed in the parallel (a) and in the normal (b) to the resonator axis. Biasing magnetic field strength: curve 1 – 1430 Oe, curve 2 – 3530 Oe.

cell showed that these elements demonstrate strong non-reciprocity of electrophysical properties. In some cases the isolation reaches ~50 dB, so the difference between energy transmission values for the “direct” and “reverse” wave passage reaches ~99.8% (Fig. 6). It must be said that at the resonant frequency the reflection does not exceed 9% in power, and at other frequencies in the operating range it is even lower. It means, that the main contribution to the transmission spectrum makes absorption, not reflection. Therefore, the reflectance spectra were not analyzed separately during the work and the corresponding data in the text of the manuscript are not given. In the future reflection losses can be reduced by matching the resonator in the waveguide.

Theoretically such resonators can be used for the design of microwave devices with magnetic field tunable nonreciprocal characteristics. However, although high isolation values were observed, direct losses in some cases (see Fig. 6) reached 10–14 dB (80%) while for the technical purposes this value must be less than 1.5 dB. Therefore with the aim to determine the possibility of obtaining of necessary characteristics of a non-reciprocal element we made the simulation of the composite resonator in the waveguide measuring cell using the *ANSYS HFSS*

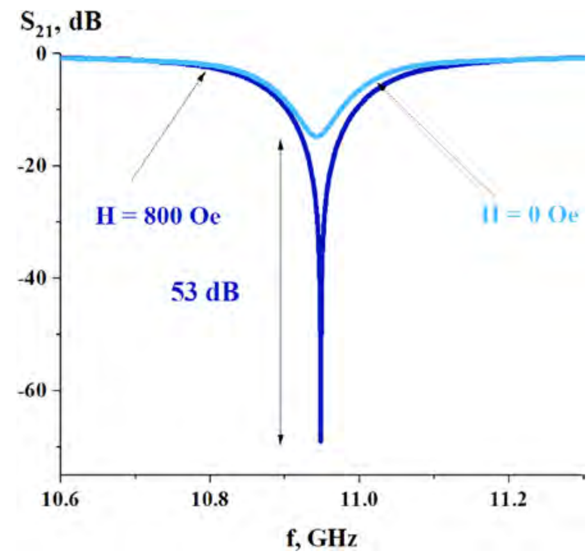

Fig. 6. Non-reciprocity of energy transmission spectra of the composite resonator with ferrite film polymerized in the magnetic field directed in the parallel to the resonator axis.

software (Fig. 7). It can be seen from Fig. 7 that due to the dielectric resonator presence the electromagnetic field energy concentrates inside the structure. It is one of the reasons of the fact that the alternate magnetic field amplitude inside the ferrite film is 8–10 times higher than it is in the rest of the waveguide volume. Therefore the resonator increases the effectiveness of interaction between the ferrite film and electromagnetic field and improves the non-reciprocity and isolation value. As a result, the simulation revealed that when the resonator is located at certain points of the waveguide cross section it is possible to achieve the direct losses less than 0.2–0.8 dB and reverse losses approaching 34 dB (Fig. 8a) at the resonant frequency of $H_{01\delta}$ eigenmode (Fig. 8). As numerical simulation showed, the optimal positions of the composite resonator in the waveguide under the mentioned conditions are near the narrow walls of the waveguide. For comparison, the transmission spectrum of a single ferrite film with the same magnetic parameters (and without dielectric resonator), set near a narrow wall, is shown on Fig. 8b. It can be seen that the transmission coefficient is also non-reciprocal, but the value of the isolation is much smaller than for the ferrite-dielectric structure. So, the non-magnetic constituent of the resonator pull the field in and the magnetic film effectiveness rises, the synergistic effect occurs.

Thus, the numerical simulation shows that if a high quality ferrite film is produced and deposited on a low-loss dielectric resonator, such composite resonant element can be truly nonreciprocal, with both high values of isolation and small values of energy losses for the wave passing in the direct direction.

4. Conclusions

Multiphase high-quality dielectric materials have been investigated. It is shown by the example of the $BaTi_4O_9$ – ZnO system that the increase of the Q-factor in multiphase systems can occur due to the appearance of an additional phase with the Q-factor higher than in the main phase. Improvement of the electrophysical properties temperature stability in multiphase systems is achieved due to the effect of volumetric thermal compensation in the presence of two phases with the different signs of dielectric constant temperature factor.

Composite resonators are developed on the basis of high-quality dielectric materials and ferromagnetic film, which are characterized by high quality factor and ability to control the resonance frequency in a magnetic field. It is shown that such resonators are characterized by the

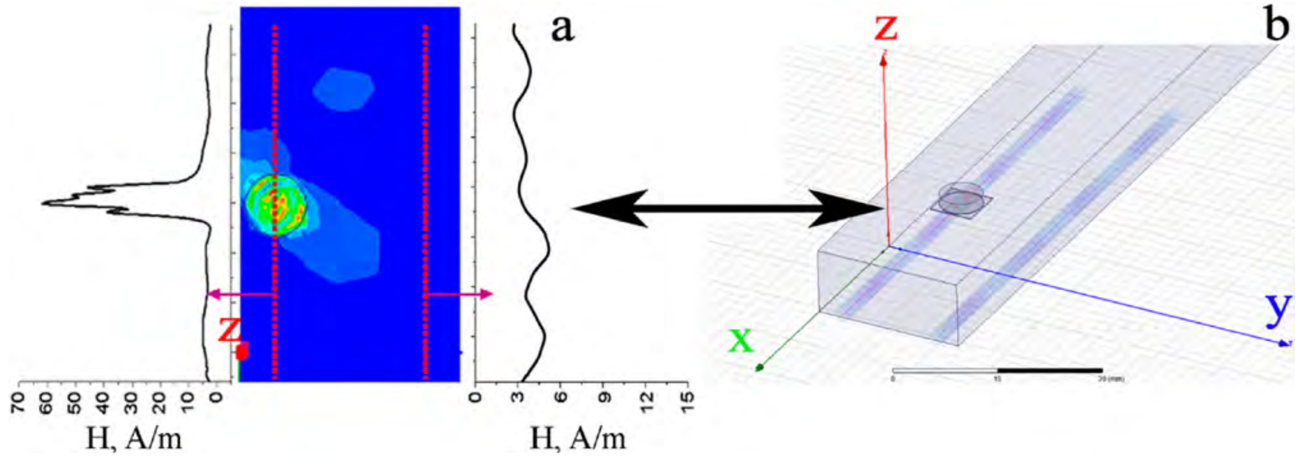

Fig. 7. Magnetic field distribution along imaginable symmetrical lines inside the measuring cell with the composite element (a); the geometry of the measuring cell model (b).

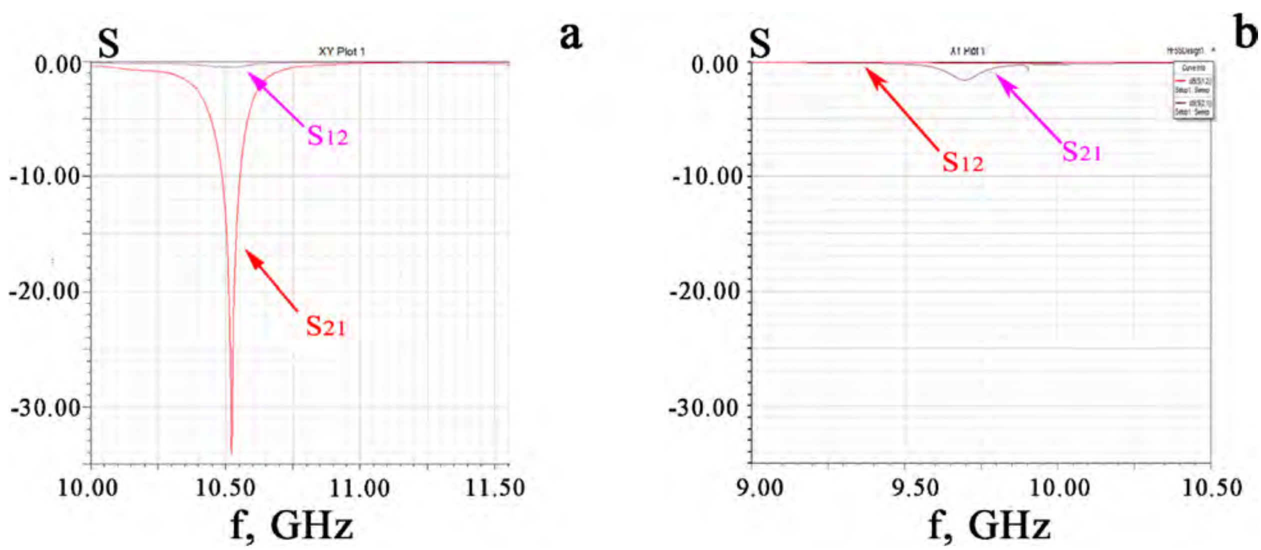

Fig. 8. Energy transmission spectra of the composite ferrite-dielectric resonator model (a) and of a single ferrit film (b) for the same value of bias magnetic field.

very high non-reciprocity of microwave energy transmission.

The obtained composite resonant elements can be used in non-reciprocal, field-controlled waveguide and microstrip components for radar stations, filter cascades, microwave isolators, superheterodyne transmitters. These resonators may also be designed for the lower frequency bands and used in GSM equipment or wireless Wi-Fi.

CRediT authorship contribution statement

Anatolii Belous: Conceptualization, Supervision. **Oleksandr Fedorchuk:** Investigation, Writing - review & editing. **Sergii Solopan:** Investigation, Visualization, Resources, Writing - original draft. **Maksym Popov:** Methodology, Formal analysis, Writing - original draft. **Igor Zavislyak:** Conceptualization, Supervision.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jmmm.2020.166691.

References

[1] A.G. Belous. High-Q microwave dielectrics. Kyiv: Naukova dumka, 2016, p. 219. ISBN: 978-966-00-1544-9.
[2] A. Belous, O. Ovchar, M. Valent, et al., Abnormal behavior of the dielectric parameters of $Ba_{6-x}Ln_{8+2x/3}Ti_{18}O_{54}$ (Ln = La-Gd) solid solutions, J. Appl. Phys. 92 (7) (2002) 3917–3922.
[3] A.G. Belous, O.V. Ovchar, O.V. Kramarenko, et al., Low-loss microwave ceramics based on non-stoichiometric perovskites $Ba(Co_1/3Nb_{2/3})O_3$ and $Ba(Zn_1/3Nb_{2/3})O_3$, 11^{th} European Meeting on Ferroelectricity, Slovenia, Bled. – Bled. (2007) 52.
[4] R. Cameron Kudsia, W.-C. Tang, Innovations in microwave filters and multiplexing networks for communications satellite systems, IEEE Trans. Microwave Theory Tech. 40 (1992) 1133.
[5] S.J. Fiedziuszko, Trends and evolution of high performance filters, Proc. IEEE 2001 MTT-S Int. Microwave Symp., WMG Workshop, (2001).
[6] J.S. Hong, Reconfigurable planar filters, IEEE Microwave Mag. 10 (6) (Oct. 2009) 73–83.
[7] R.R. Mansour, IEEE Microwave Mag. 10 (2009) 84–98.
[8] Y.M. Poplavko, Y.V. Prokopenko, V.I. Molchanov, A. Dogan, IEEE Trans. MTT 49 (2001) 1020–1026.
[9] O.G. Vendik, E.K. Hollmann, A.B. Kozyrev, A.M. Prudan, Ferroelectric tuning of planar and bulk microwave devices, J. Supercond. 12 (1999) 325–338.
[10] K.D. Soloviova, O.P. Fedorchuk, M.O. Popov, I.V. Zavysliak, A.G. Bilous, Development the composite magnetically heterostructures based on "thick film of

spinel nickel ferrite — dielectric resonator a-Al_2O_3, Ukrainian Chem. J. 82 (5) (2016) 10–14.
[11] V.G. Tsykalov, V.M. Pashkov, V.P. Bovtun, “The batch for a manufacturing of the temperature stable ceramic dielectric” [in russian], USSR patent 3286870, 1983.
[12] Yu.I. Gornikov, Z.Ya. Makarova, A.G. Belous, L.G. Gavrilova, V.M. Paskov, V.P. Chalyi, Sov. Prog. Chem. 12 (50) (1984) 1243–1245.
[13] A.I. Tovstolytkin, M.M. Kulyk, V.M. Kalita, S.M. Ryabchenko, V.O. Zamorskyi, O.P. Fedorchuk, S.O. Solopan, A.G. Belous, Nickel-zinc spinel nanoferrites: magnetic characterization and prospects of the use in self-controlled magnetic hyperthermia, J. Magn. Magn. Mater. (2018), https://doi.org/10.1016/j.jmmm.2018.10.075.
[14] L. Barbieri, A. Ferrari, I. Lancellotti, C. Leonelli, J.M. Rincyn, J. Am. Ceram. Soc. 83 (2000) 2515.
[15] YuI. Gornikov, Z.Ya Makarova, A.G. Belous, Sov. Pro. Chem. 12 (50) (1984) 1243–1245.
[16] R.S. Roth, C.J. Rawu, C.G. Lindsay, W. Wong-Ng, J. Solid State Chem 104 (1993) 99–118.
[17] L.F. Chen, C.K. Ong, C.P. Neo, V.V. Varadan, Vijay K. Varadan, Microwave Electronics: Measurement and Materials Characterization, John Wiley & Sons, 2004 DOI: 10.1002/0470020466.
[18] M.A. Popov. Ferromagnetic resonance in a single crystal of iron borate and magnetic field tuning of hybrid oscillations in a composite structure with a dielectric: Experiment and theory / M. A. Popov, I. V. Zavislyak, H. L. Chumak, M. B. Strugatsky, S. V. Yagupov, G. Srinivasan // J. of Appl. Phys., 2015, vol. 118, no. 1. -P. 013903-1- 013903-9, DOI: 10.1063/1.4923413.
[19] A.G. Gurevich, G.A. Melkov, Magnetization Oscillations and Waves, CRC Press, New York, 1996.
[20] B. Lax, K. Button. Microwave ferrites and ferrimagnetics [Russian translation, red. A.G. Gurevich], Mir, Moscow, p. 676, 1965.
[21] Raul Valenzuela, The temperature behavior of resonant and non-resonant microwave absorption in Ni-Zn ferrites, Electromagnetic Waves (2011), https://doi.org/10.5772/16508.
[22] Yu Sitidze, Kh Sato, Ferrites [Russian translation], Mir, Moscow (1964).
[23] Qian Liu, Lu. Li, Jian-Ping Zhou, Xiao-Ming Chen, Xiao-Bing Bian, Peng Liu, J. Ceramic Process. Res. 13 (2) (2012) 110–116.