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Cold pressing of ferroelectric-ferromagnetic layered composites for nonlinear forming lines of high-voltage impulse generators

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The results of investigation on synthesis of the layered composites "ferromagnetic-ferroelectric " for usage as an active dielectric of the impulse current and voltage generators' forming lines are presented. The structure of composites with simultaneous change of their effective permittivity and permeability to achieve almost constant wave impedance of the forming line at electromagnetic waves propagation are proposed. The technological methods, which provide obtaining the layered composites samples by cold pressing, are described.

Keywords: layered ferroelectric-ferromagnetic composites, non-linear electric and magnetic properties, cold pressing method, forming lines, pulse generators of currents and voltages.

Представлены результаты исследований по синтезу сегнетомагнитных композитов, предназначенных для использования в качестве активного диэлектрика формирующих линий генераторов импульсных токов и напряжений. Предложена структура композитов, обеспечивающих синхронное изменение их эффективных диэлектрической и магнитной проницаемостей с целью достижения изоимпедансности (постоянства волнового сопротивления при взаимодействии нелинейного композита с электромагнитным полем). Описаны технологические приемы, позволившие изготовить опытные образцы слоистых композитов методом холодного прессования.

Холодне пресування шаруватих сегнетомагнітних композитів для нелінійних формуючих ліній високовольтних імпульсних генераторів. *О.Л.Резинкін, М.М.Резинкіна, О.Г.Гриб, В.І.Ревуцький.*

Представлено результати досліджень з синтезу сегнетомагнітних композитів, призначених для використання в якості активного діелектрика формуючих ліній генераторів імпульсних струмів і напруг. Запропоновано структуру композитів, що забезпечує синхронну зміну їх ефективних діелектричної та магнітної проникності з метою досягнення ізоімпедансності (сталості хвильового опору при взаємодії нелінійного композиту з електромагнітним полем). Описано технологічні прийоми, що дозволили виготовити дослідні зразки шаруватих композитів методом холодного пресування.

1. Introduction

One of the important tasks of strong electric and magnetic fields technique is creation of power impulse generators of voltages and currents with short risetime [1]. Such generators are necessary, in particular for carrying out experimental research in plasma physics, for realization of the modern electrical physical technologies, as well as for simulation the effects of influence of natural and man-made electromagnetic disturbances on technical objects. Nonlinearity of dielectric and ferromagnetic media's parameters can be used for such generators creation. Thus, usage of sharpeners on ferrites provides obtaining current pulses with the amplitude up to hundreds of amperes with submicrosecond fronts [2].

Deformation of electromagnetic waves profile occurs at their propagation in the media with nonlinear parameters. This deformation can lead to electromagnetic shock waves appearance and be accompanied by shortening of rise time of the electric and magnetic fields impulses [3].

As dielectric or magnetic permeability of the nonlinear medium under influence of the strong electromagnetic fields does not remain constant, wave impedance of the sources of pulsed currents and voltages, which contain a medium with nonlinear properties of only permeability or permittivity, changes in time. The inability of matching of changing wave impedance of such impulse sources with a constant load causes incomplete transfer of power to the load. For this reason, the task of creation and study of the medium in which both permittivity and permeability would change simultaneously, making the wave impedance close to a constant is of current interest. This improves the matching of the wave resistance of pulse generators using the electromagnetic shock waves with the load [4].

Polycrystalline mediums having simultaneously ferroelectric and ferrimagnetic properties have been described elsewhere [5]. For such mediums it was observed an influence of the magnetic field strength not only on the permeability, but also on the permittivity as well as influence of the electric field both on the permittivity and the permeability. However, at creating the active dielectric medium for forming lines (FL) of impulse current and voltage generators usage of such mediums causes the considerable difficulties. To ensure reliable operation of the nonlinear forming lines it is necessary to synthesize the medium in which the permittivity and permeability change simultaneously, but independently.

The aim of this work is to develop methods for obtaining the composites samples with nonlinear permittivity and permeability and suitable for creating forming lines of impulse current and voltage generators with wave impedance close to constant.

2. Experimental

2.1. Assessment of equivalent parameters of ferroelectric - ferromagnetic composites

To obtain reliable data on the operation of the electrical devices using non-linear electric and magnetic properties of medium, physical and mathematical modeling of the electrical physical processes may be used.

Calculations of the transients in the nonlinear forming lines at electromagnetic waves propagation allow giving the recommendations on the choice of degree of the non-linearity of permittivity and permeability of working medium required to achieve assigned wave-front deformation, the oscillations level and the other current and voltage output parameters. Considering the complexity of these tasks, they are usually solved by presentation a forming line with distributed parameters as an artificial transmission line with lumped electrical parameters [6]. However, such an approach does not take into account a number of peculiarities at the electromagnetic wave propagation caused by 3D configuration of the considered system elements.

Delay of propagation along the line of the wave with a short rise time should be taking into account at calculation of the considered electromagnetic processes. Formulation of the problem through the potentials (scalar electric and vector magnetic) reduces the number of equations to be solved by one-third, which is important in three-dimensional approach. Solution of the considered problem analytically is impossible, so the numerical methods were used for the calculations. Taking into account that most of the borders between the media in the investigated lines are flat, numerical method of the final integration technique can be applied for solution [7–10]. Such a method involves rectangular computational grid application to the considered area and integration of the Maxwell's equations over the volumes or cross-sections of the cells. This approach allows solution of the equations using the law of charge conservation or the total current law. As a result, it is not necessary to use additional equations describing conditions on the boundaries between the media, as they are taken into account automatically.

Information about equivalent dielectric and magnetic parameters of the used composite media is required for mathematical and physical modeling of the investigated electromagnetic processes.

For example, as the ferromagnetic fraction in the "ferroelectric-ferromagnetic" composite, ferrite Epcos K1 with a binder of calcium stearate can be used. Application of relations obtained in [11], which connect equivalent parameters of the ferromagnetic matrix mixture with concentrations and properties of the included materials allows evaluating of the equivalent magnetic permeability values of such layers. A method has been described in [11] for calculation of μ_e — equivalent magnetic permeability of the ferromagnetic composite mixture having the relative magnetic permeability μ_1, and a filler having the relative magnetic permeability μ_2:

$$\mu_e = 0.5 \cdot (1 - 2p_\mu)(\mu_2 - \tilde{\mu}_1) + $$
$$+ 0.5\sqrt{(1 - 2p_\mu)^2(\mu_2 - \tilde{\mu}_1)^2 + 4\mu_2\tilde{\mu}_1}, \quad (1)$$

where $\tilde{\mu}_1$ is determined from the dependence $\mu_1(H)$, H is strength of applied magnetic field, p_μ is volume concentration of ferromagnetic material in the composite, $\mu_2 = 1$.

This expression for μ_e is used for recalculation of the averaged over the ferromagnetic inclusions volumes magnetic field H_{av} and $\tilde{\mu}_1$ recalculation from the curve $\mu_1(H)$ by obtained H_{av} level [11].

Three-component system $BaO–SrO–TiO_2$ with dopands [12], having the high non-linear characteristics, can be used as the ferroelectric fraction in the "ferroelectric-ferromagnetic" composite. Such layers consist of dispersed ferroelectric ceramic, and may be represented as statistical mixtures of ferroelectric with relative permittivity ε_1 and air with $\varepsilon_2 = 1$. In [13] it is described a technique for calculation of ε_e — the equivalent permittivity of such mixtures when presenting the powder particles as spheres. For $\varepsilon_2 << \varepsilon_1$ and $\varepsilon_2 << \varepsilon_e$:

$$\varepsilon_e \approx \varepsilon_1(1.5p_\varepsilon - 0.5), \quad (2)$$

where p_ε is volume concentration of ferromagnetic material in the composite.

Creation of the non-linear medium with close to equal wave impedance requires synchronous saturation of the electric and magnetic field inductions in the "ferroelectric-ferromagnetic" composite. For this, the ratio $\sqrt{(\mu_e)/\varepsilon_e)}$ (where μ_e and ε_e are relative equivalent of permeability and permittivity of ferromagnetic and ferroelectric layers, respectively) should remain close to the constant. Using the experimental relation between D electrical induction and E-electric field strength (see for example [14]), it is possible to choose a section on the curve $B(H)$ (where B is magnetic induction) with the same curvature as in the used range $D(E)$ and obtain the desired range of the magnetic field strength saturation values in the ferromagnetic layers from the relation between the electric and magnetic fields strength: $H_{S\mu} = K_G \times E_{S\varepsilon}\sqrt{(\varepsilon_{S\varepsilon})/\mu_{S\mu})}$ (where K_G is a coefficient depending on the geometry of FL; $S\varepsilon$, $S\mu$ are indices relating to the considered range changes of the electric and magnetic fields, respectively). Varying of the levels of volume concentrations p_μ and p_ε (see (1,2)), as well as K_G level by means of changing the FL geometry, for example, the ratios between width of the electrodes and thickness of the "ferroelectric-ferromagnetic" composite, can provide the required $H_{S\mu}$ and $\mu_{S\mu}$ levels, assigning the specific $E_{S\varepsilon}$ and $\varepsilon_{S\varepsilon}$ levels.

2.2. Elaboration of the structure of ferroelectric-ferromagnetic composites

Experiments on cold pressing of samples with different ratios of ferroelectric and ferromagnetic fractions of the ferroelectric-ferromagnetic composites were carried out for elaboration of technological methods of their synthesis and investigation of their electrical physical properties. At such experiments, the samples of uniform statistical mixtures of dispersed ferroelectric and magnetic dielectrics were synthesized. A number of homogeneous ferroelectric-ferromagnetic mixtures of powder ferroelectrics based on barium strontium titanate, carbonyl iron and polymer binder (PTFE, emulsion PVC, calcium stearate) were carried out. Nonlinearity of the permittivity and permeability of these mixtures depend on the concentration of contained ferroelectric and ferromagnetic components. A series of experiments to determine the properties of the obtained homogeneous compounds was performed. These investigations have shown that to choose concentration of the components of such mixtures in such a way, that the resulting composite medium has almost constant impedance, is not be possible. The matter is the conductive inclusions (carbonyl iron) at the concentration necessary to achieve a pronounced nonlinearity of the magnetic properties of the composite causes appearance of percolation paths connecting the forming lines electrodes and increasing of the synthesized active medium conductivity. If magneto-dielectrics (e.g., ferrites) are used as the ferromagnetic phase in statistical mixture of the composite, unacceptable decrease of the dielectric strength of the statistical mixture occurs at concentrations required to achieve almost constant impedance condition.

It was found as a result of the experimental investigations of electrical physical properties of the "ferroelectric-ferromagnetic" composites that to provide their adequate electrical strength and to prevent flow-through currents through percolation conductive paths, which can be formed by ferromagnetic (or ferrimagnetic) particles of the working medium, the structure of the composite should be layered with the layers oriented transversely to the power lines of the applied electric field.

Fig. 1 shows a cross section of a nonlinear forming line with the ferroelectric-ferromagnetic layered composite as an active dielectric for generation power shock waves and having the electromagnetic characteristic impedance close to a constant value. It may be assumed that vectors of electric and magnetic field strengths are perpendicular to each other at the electromagnetic waves propagation for this system. The electric field in the most part of volume is concentrated between electrodes *1* and directed perpendicular to ferromagnetic composite layers *2*, *3* and the magnetic field is directed along these layers. For clarity, Fig. 1 is shown not to scale. The thickness of the ferroelectric-ferromagnetic layers of the composite FL should be at least an order less than the electrodes width. Such a ratio provides close to the uniform spatial structure of both electric and magnetic components at the electromagnetic wave propagating along the forming line, which is necessary for uniform sharpening of its front. The equivalent permittivity and permeability can be assessed in accordance with (1, 2).

Reliable insulation of high voltage electrodes of the forming line was achieved for the multilayered ferroelectric-ferromagnetic composite media, synthesized by cold pressing of the ferroelectric and ferrite powders with a polymer filler. As such a binding filler, the suspension of polyvinylchloride PVX-C-6388-ZH and calcium stearate was used.

Cold pressing is a relatively easy way for implementation technology of the non-linear layered composite materials formation. This technology provides a possibility of composites creation from the ferroelectric particles and carbonyl iron powder or ferrite mixtures with the polymer filler, followed the layer by layer compressing them into a mold. The high degree of the used ferroceramics cryogenic gas dynamic grinding allows obtaining of the dense layers with the fine-grained structure, small polarization domains and minimized polarization duration at impulse electromagnetic fields application.

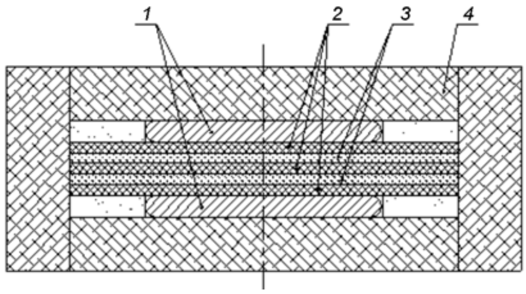
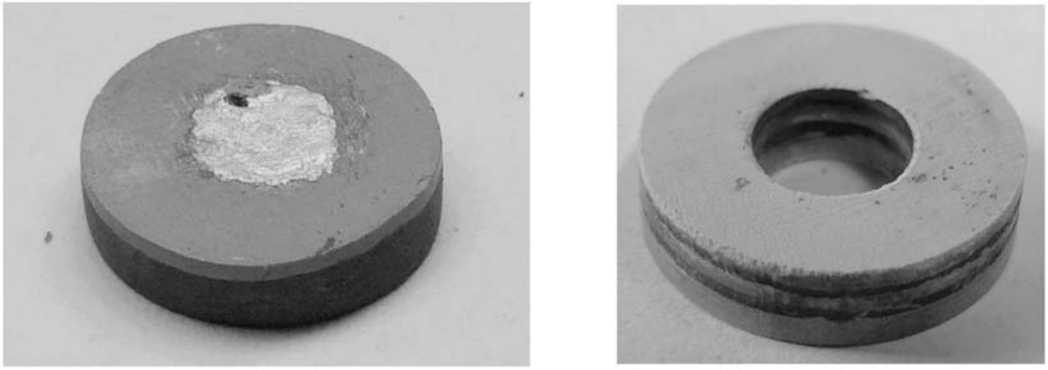


Fig. 1. Cross-section of a layered forming line. *1* — are electrodes; *2* — are ferroelectric layers; *3* — are ferromagnetic layers; *4* — closed magnetic circuit (ferrite cores from Epcos K1 material).

In the process of a the composite forming, powders of a solid solution of barium titanate and soft magnetic ferrite were mixed with the polymeric binder. Shaping of the experimental samples composites was carried out by cold pressing at a static pressure. The layered composite samples were produced in the form of cylinders up to 2 mm of height with the square of deposited on their base circular silver electrodes of 50 mm^2 (Fig. 2a), and circular rings of 12×5×4 mm (Fig. 2b) with the axes normal to the layers of ferroelectrics and ferromagnetics. Electric and magnetic parameters of the synthesized layered medium in electric and magnetic fields were determined on the experimental samples.

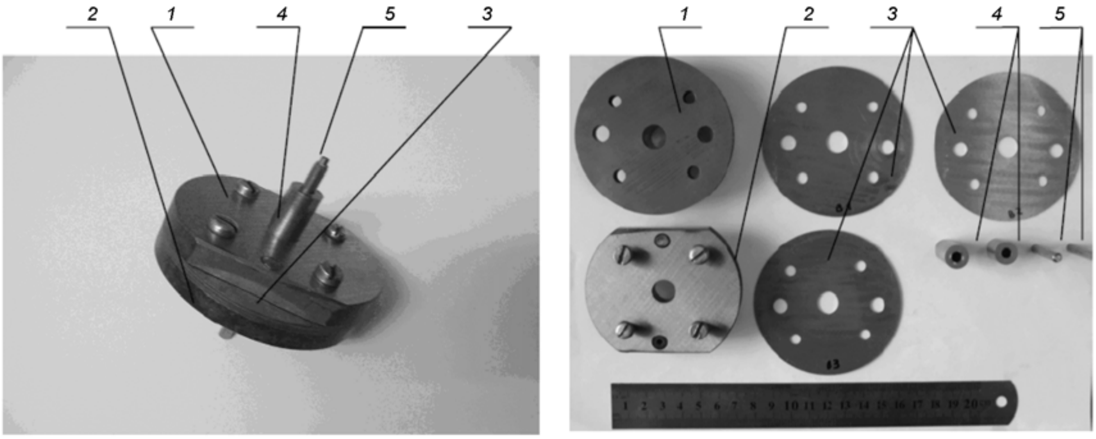
The diameter of the experimental samples is several times bigger than their height, which made it possible to put the ferroelectric-ferromagnetic samples in planar electrode system and carry investigations of their polarization in the close to homogeneous electric field, which direction was perpendicular to the layers of the composite. The annular shape of these samples has allowed to apply to them windings of copper wire, forming in the ferroelectric-ferromagnetic material the magnetic field with strength parallel to the direction of its layers, and measuring windings to determine the magnetic field induction.

Technological rigging used for the composite sample shaping in the form of cylinders and rings is shown in Fig. 3. The pressing was carried out inside the de-



a) b)

Fig. 2. Samples of layered ferroelectric-ferromagnetic composites with alternated layers of ferroelectric and ferromagnetic: a) cylindrical; b) toroidal.



a) b)

Fig. 3. Mold– a) mold assembly; b) mold components: *1* and *2* — are the upper and lower parts of the demountable mold; *3* — are flat plates; *4* — are punches; *5* — are axial insertion into punches for toroidal samples pressing.

mountable mold 1, 2, by compression of powder mixtures of active dielectrics with the binder by two punches moving towards each other by means of a hydraulic press. The pressing pressure was 14 MPa, maintaining the same pressure during from 1 to 3 min. at room temperature. The ferroelectric and ferromagnetic layers were recompacted alternately to cylindrical or toroidal samples after installation in the mold of the flat plates *3* (see Fig. 3) corresponding to their thickness and filling the necessary amount of powder. Tubular punches *3* with axial inserts *4* were used for the toroidal samples compression. By varying the ratio of non-linear components of the mixture and the filler, acceptable mechanical strength of the samples was achieved. Fig. 4 shows SEM micrographs of the fracture of the composite sample manufactured by the described technology.

The cylindrical test samples were used for investigation of polarization parameters of the manufactured ferroelectric-ferromagnetic layered composites. The samples, containing dispersed ferroelectric based on solid solutions of $(Ba_{1-x}Sr_x)TiO_3$ doped by zirconium were investigated at various electric field strengths and temperatures close to the Curie point for the used ferroelectric.

The toroidal samples were used in the experiments on magnetization of the synthesized composites investigations. For this, two coils from insulated copper wire were applied on their surface. One of the coils was used for excitation of the sample in the magnetic field with amplitude sufficient for manifestation of the nonlinear dependence of $B(H)$ and to achieve the magnetic field induction saturation level B_s. The other winding allowed measurement of the magnitude of induced EMF and thus to determine the value of B as time integral.

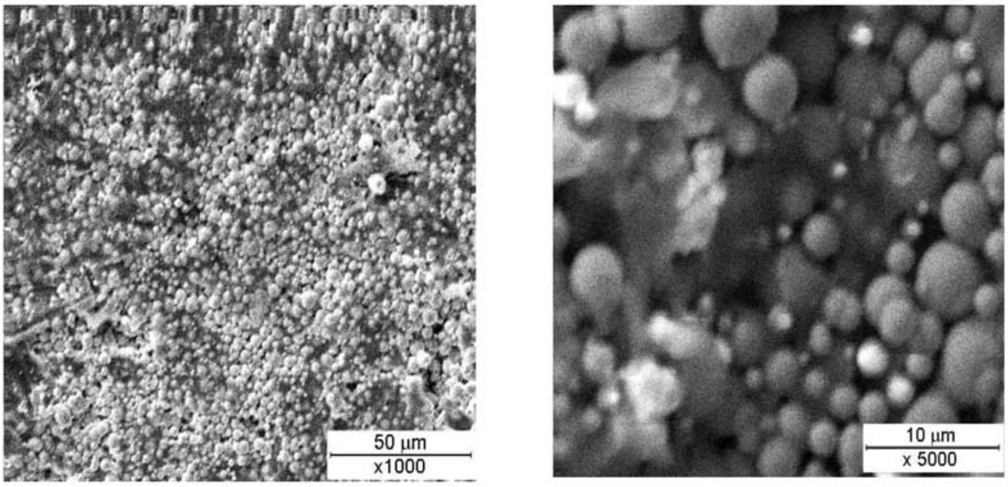


Fig. 4. SEM micrographs of fracture surface of the composite sample.

Experimental investigations of the electrical physical characteristics of the composites were carried out by oscilloscopic methods in sinusoidal and pulsed electromagnetic fields.

Investigations of the composites dielectric characteristics were carried out using the Sawyer-Tower method [15] at 25–70°C (in more details see [14]). It is known, that the nonlinear properties of ferroelectric are most pronounced at temperatures close to the Curie point. However, the optimum in terms of combination of the high nonlinearity dependence $D(E)$ and minimization of the relaxation losses in the ferroelectric phase is a temperature above the Curie point at 2–3 degrees. It should also be noted that in the ferroelectric composite the magnitude of the electric displacement approaches saturation level D_s at the electric field close to the electric strength. This causes the necessity to take measures to prevent electrical breakdown of the ferroelectric-ferromagnetic composites. Electric strength of the ferroelectric- ferromagnetic composites may be improved substantially with the help of their vacuum impregnation by liquid dielectrics or processing the surfaces by electrical insulating varnish. Impregnation of the obtained samples by T-1500 or BBG oil increased their dielectric strength in 1.5–2 times (as compared with this characteristic measured for untreated samples), depending on the rate of increase of the applied electric field strength. To prevent electrical breakdown on the surface and corona discharges occurrence at the electrode edges, investigated samples were placed in dry capacitor oil at the high voltage experiments.

The magnetic characteristics of the ferroelectric-ferromagnetic samples were determined by synchronous recording of current waveforms of the excitation coil and integrated over time voltage on the measuring winding applied to the samples of toroidal ferroelectric-ferromagnetic composites.

Analysis of forms for the obtained dependences of electric induction on electric field strength as well as the magnetic induction versus the magnetic field strength indicate the possibility of practical usage of the manufactured composites as a working medium of the high voltage nonlinear FL with constant wave impedance.

3. Conclusions

To select the content of the metal ferromagnetic powder (carbonyl iron) as a ferromagnetic component of isotropic ferroelectric-ferromagnetic mixture so that the resulting medium would be equal-impedance is not possible. This is explained by the fact that conducting inclusions concentration required to achieve the expressed nonlinearity of the mixture magnetic properties causes the appearance of conductive percolation paths connecting the forming line electrodes, which causes unacceptable rise of the conductivity or reduction of electrical strength of the synthesized active medium.

To achieve of the working medium wave impedance close to constant, the structure of forming lines on the ferroelectric-ferromagnetic composite should be layered. In such a composite layers with ferromagnetic properties should be alternated with layers having ferroelectric properties, and the interfaces of ferroelectric and ferromagnetic layers must be parallel to the surfaces of the FL electrodes. It is possible in this case to achieve simultaneous changes of equivalent permittivity and permeability of the ferroelectric-ferromagnetic composite and provide necessary insulation of the forming line electrodes. Cold pressing of ferroelectric and ferrite powders with the binder of PTFE, emulsion PVC or calcium stearate may be used for synthesis of the ferroelectric-ferromagnetic composite with the wave impedance close to the constant.

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