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# Microwave properties of composite with high content of carbonyl iron microparticles of different grades

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The results of experiments on obtaining a structural radio material promising for protecting electronic equipment with an increased (more than 80 mass%) content of carbonyl iron micropowders of various grades in a silicone compound are presented. The measured values of complex magnetic permeability in the frequency range of 0.1−6 GHz significantly exceed the known values with a lower content of micropowders. The highest absorption coefficients are observed in the created composite with a content of 88.9 mass% of grade P10 micropowder.

**Keywords:** radio absorption, carbonyl iron, complex magnetic and permittivity, hysteresis.

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The development of radio-absorbing materials for protection of electronic equipment and suppression of reflection of electromagnetic waves in various frequency bands is a relevant objective [1]. Microwave absorbance of various particles with magnetic properties (both individual particles and their combinations with polymers and conducting and dielectric nano- and microstructures) and multilayer coatings has been studied extensively [2–5]. The combination of high saturation magnetization and electrical polarizability makes carbonyl iron microparticles (CIP) particularly promising. Their radio-absorbing properties have been studied for a long time [6–9].

It is known [10] that complex permittivity and permeability are the key parameters of a radio-absorbing material; therefore, their determination may reveal new approaches to enhancing the radio-absorbing properties of a composite. The complex permittivity and permeability of composite materials made from a polymer binder based on phosphated CIP (R100F-2) were measured in [11] within the frequency range of 0.1–39 GHz. Their dependences on filler concentration (3.3–40.3 vol.%) in composites were studied, and it was found that the static permittivity and permeability of composites grow logarithmically with increasing filler percentage; the imaginary part of permeability increased up to a value of 1.2. The complex permeability of the Fe–$SiO_2$+ paraffin matrix composite with 20 vol.% of filler at frequencies up to 20 GHz was determined in [12]; its imaginary part did not exceed 0.5.

In general, it was concluded that CIP in composites offers fine potential for microwave absorption. Further research is needed to develop efficient absorbers tailored to specific frequency ranges and practical applications.

One approach to enhancing the absorbance of a composite is to increase the CIP content. A composite of chlorinated polyethylene (CPE) and CIP, which was produced by extruding the mixture and passing it through two rollers, featured reduced reflection losses (the CIP:CPE weight ratio varied from 81 to 94 mass% of CIP) within the 2–18 GHz range. However, the complex permeability indices of composites have not been measured [13]. The absorbance of a composite produced under a pressure of 15.5 MPa and consisting of silicone rubber and 50−80 mass% of CIP increased as the CIP concentration increased from 50 to 70 mass%, but remained unchanged at 80 mass% [14]. The absorption properties of composites with high CIP concentrations were reviewed in 2021 in [15], and it was concluded that the optimum percentage of CIP providing a high absorbance needs to be determined. Samples of epoxy paint for coating military equipment with 75−80 mass% of CIP were prepared using a stirrer and applied to a metal plate. Radar measurements of microwave attenuation in these samples were compared with measurements for epoxy paint without CIP, and it was found that the samples with 75 mass% provide stronger attenuation than those with 80 mass% [8]. The results of theoretical calculations for a CIP concentration of 100 % (intrinsic value) performed using the dispersion relation [11] indicate that high complex permeability values may be observed in composites with a high CIP content. We believe that the radio absorbance of composites with a high CIP content is understudied, and the experimentally obtained values of complex permittivity and permeability of composites often fall short of the levels feasible for composites with CIP. In addition, comparative data on the radio absorbance of composites with different CIP grades remain unavailable.

This is likely attributable to the fabrication of electrically conductive samples, which leads to an impedance mismatch and, consequently, reflection of electromagnetic waves at insufficient binder concentrations in the composite, and to technological difficulties arising in the process of fabrication

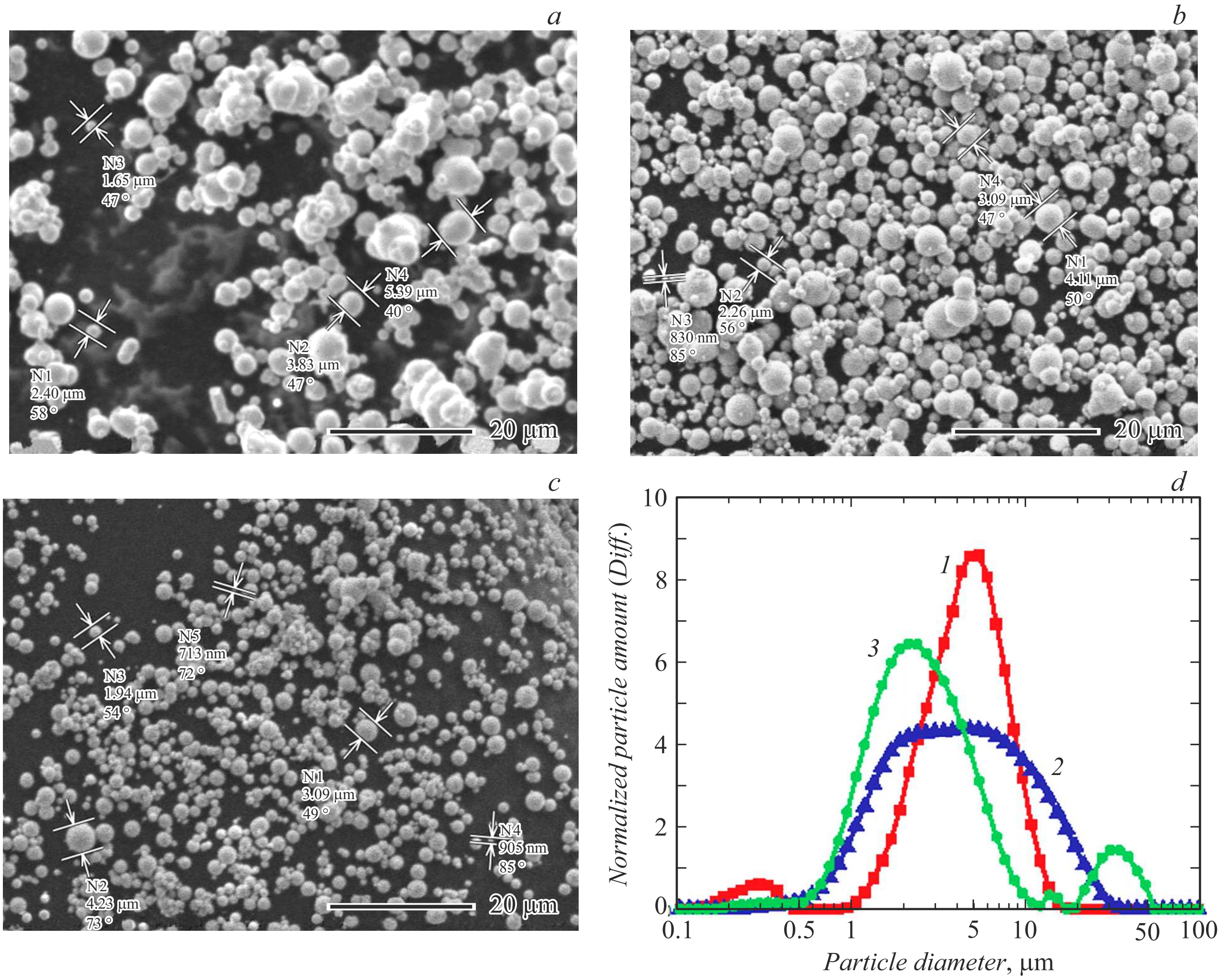


**Figure 1.** SEM images of carbonyl iron micropowders R10 (*a*), R20 (*b*), and R100F-2 (*c*). *d* — Distribution of microparticles by size: R10 (*1*), R20 (*2*), and R100-F2 (*3*).

of composites with high (more than 80 mass%) CIP concentrations.

Thus, the aim of the present study is to produce and examine composites with a high (more than 80 mass%) concentration of CIP of various grades in silicone compound for the purpose of identifying viable approaches to fabrication of a material with increased microwave absorbance. With this end in view, the sizes and shapes of microparticles were determined and compared; the corresponding composites were produced; their reflection, transmission, and absorption coefficients were determined; and their complex permittivity and permeability were measured within the frequency range of 0.1−6 GHz. Silicone compound was chosen as a dielectric material for the following reasons: technological simplicity of mixing of carbonyl iron micropowder in a liquid form prior to polymerization and high plasticity of the formed compound. The indicated frequency range was investigated due to the high current demand for materials with low coefficients of reflection of electromagnetic radiation in radio-electronic equipment generating microwave radiation.

The efficiency of interaction of microwave electromagnetic radiation with composite materials (transmission, reflection, absorption) was examined using a „Panorama“ P4226 (Mikran, Russia) vector network analyzer (VNA). Full two-port calibration of the measuring path with a set of SOLT standards with an intermediate frequency (1 kHz) filter was performed.

The composites were produced by adding two-component tin-based silicone compound SilcoTin 25 (Khimsnabkompozit, Russia) to a powder of radio-technical carbonyl iron of three different widely used grades: R10, R20, and phosphated R100F-2 CIP (OAO Sintez, Russia). Following mechanical mixing, the samples were introduced into a coaxial measurement cell with a central core diameter of 3.05 mm and an outer electrode diameter of 7 mm. The sizes and shapes of R10, R20, and R100F-2 carbonyl iron particles were determined using a JCM-6000 (JEOL Ltd, Japan) scanning electron microscope (SEM; Figs. 1, *a*−*c*).

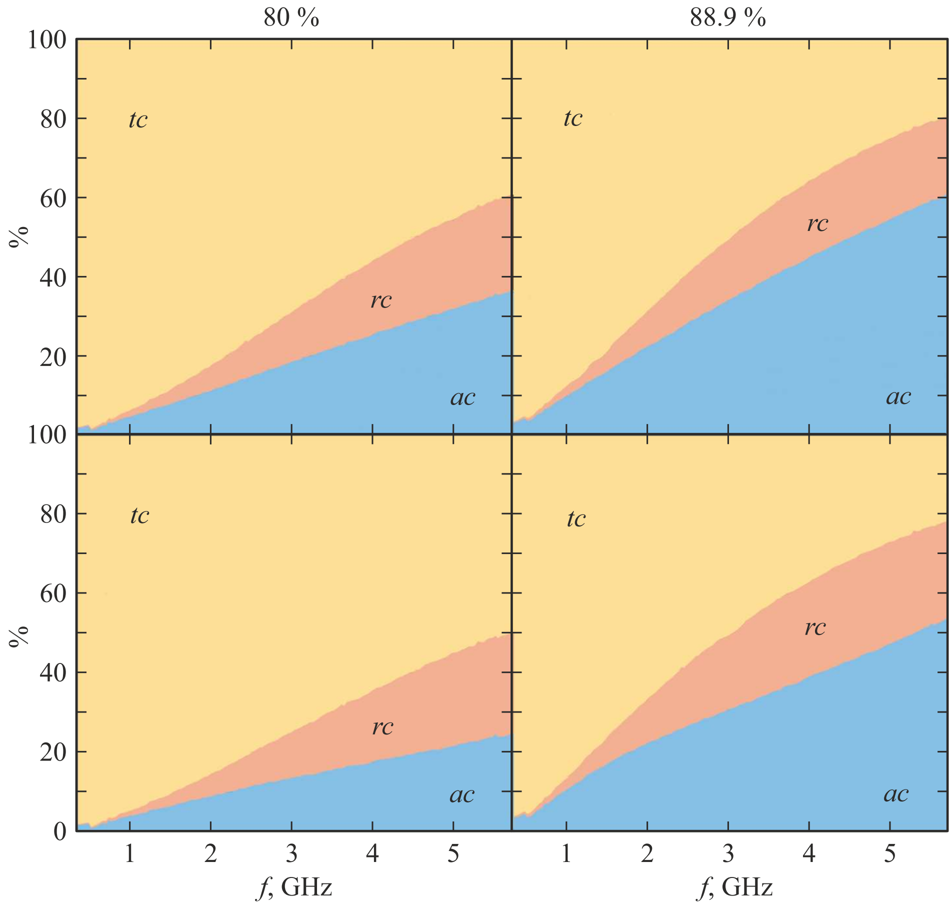


**Figure 2.** Coefficients of reflection, absorption, and transmission of electromagnetic radiation for silicone matrix SilcoTin 25 with micropowders R10 (upper row) and R20 (lower row) with different percentages of carbonyl iron filler; $ac$, $rc$, and $tc$ denote the areas corresponding to absorption, reflection, and transmission of microwave radiation.

The average diameter of microparticles measured this way was as follows: R10 — 2–4 $\mu$m, R20 — 1–3 $\mu$m, and R100F-2 — 1–2 $\mu$m; particles were spherical in shape. The distributions of microparticles by size for micropowders in distilled water were determined by a SALD-7500 particle size analyzer (Shimadzu, Japan) and were bimodal with an average size of 3.8 and 2.7 $\mu$m (R10 and R100-F2, respectively) or had a single flatter maximum with an average size of 3.8 $\mu$m (R20; see Fig. 1, $d$). The results of X-ray diffraction analysis revealed that all three carbonyl iron micropowders consisted of alpha iron. The viscous composition was introduced into a press mold and subjected to compression under a pressure of 10 MPa. The studied composite samples were cut out with a punch from the obtained composite and had the form of a flat ring with an internal diameter of 3 mm and an external diameter of 6 mm. Their thickness varied within the range of $1.9 \pm 0.2$ mm. Measurements with a DT-830B (Wenzhou Tosun Electric Co., China) digital multimeter demonstrated that the samples were poor conductors of electric current. Their resistance was no lower than 1 M$\Omega$ at a distance of 1 mm between the measuring probes.

The results of experimental studies of the interaction of microwave radiation with composites containing 80 and 88.9 mass% of CIP are presented in Figs. 2 and 3.

The absorption coefficient of all samples increases with increasing filler content (starting from 80 mass%), while the reflection coefficient remains virtually unchanged. A particularly significant absorbance enhancement (up to 60 % at a frequency of 6 GHz in the composite with 88.9 mass% of filler) is observed for the composite with R10 powder / silicone compound ratios of 4:1, 6:1, and 8:1 (80, 85.7, and 88.9 mass% of CIP); therefore, the properties of exactly this composite are discussed below. The absorption, reflection. and transmission coefficients lie between the data for 80 and 88.9 mass% of CIP.

The permeability and permittivity were determined using the proprietary software [16] for calculation of frequency dependences of the complex permeability and permittivity of materials based on the scattering parameters for traveling power waves ($S$-parameters) measured by the VNA. The results of permeability calculation for the composite with R10 are presented in Fig. 3. These measurement data were smoothed with a five-point moving average. The error of

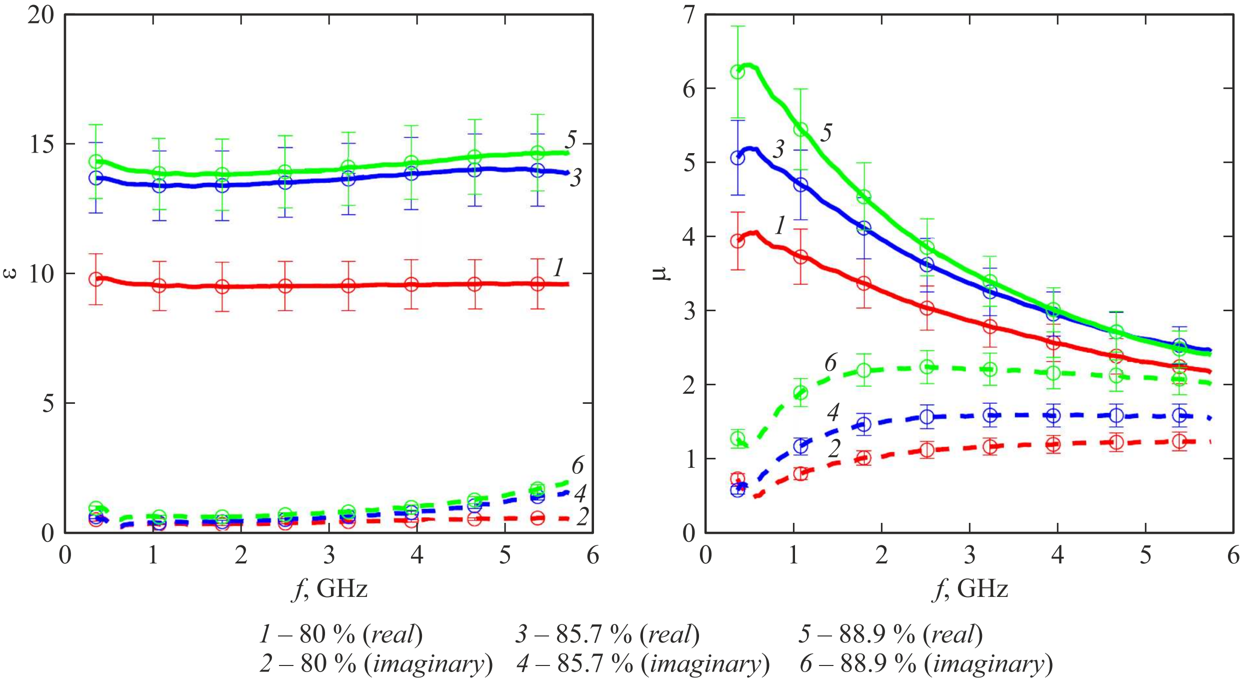


**Figure 3.** Complex permittivity $\varepsilon$ and permeability $\mu$ for composite materials with silicone matrix SilcoTin 25 containing 80, 85.7, and 88.9 mass% of carbonyl iron micropowder R10.

calculation in the program did not exceed 2 %; however, the overall error of permeability and permittivity determination was greater due to differences in roughness, variation of sample sizes, and nonuniform distributions of particles in the composite. Therefore, it was estimated by deviations from the moving average based on the measurement data for 3–5 samples with the same filler, thickness, and CIP grade and did not exceed 10 %. The permeability and permittivity values for 85.7 mass% of CIP in the composite lie between the curves for 80 and 88.9 mass% of filler. It can be seen that the imaginary part of complex permeability depends on the CIP content. The values of imaginary part of permeability of the composites obtained in the present study are approximately 2 times higher than the values for composites containing CIP with lower concentrations of 40.3 vol.% ($\sim$ 52.9 mass%) and 43.7 vol.% ($\sim$ 69.7 mass%), which were measured in [11,12], and with a high CIP concentration of 70 mass% [17] and 80 mass% [14]. The approximate conversion to mass percent was performed with account for the density of the polymer silicone binder ($\sim 1.2\,\mathrm{g/cm^3}$) and the bulk density of CIP ($\sim 2.0\,\mathrm{g/cm^3}$). The imaginary part of permeability also increases markedly in the composite with silicone compound and R10 powder in the series of mass CIP concentrations of 4:1, 6:1, and 8:1 (80, 85.7, and 88.9 mass% of carbonyl iron). The validity of the obtained data is verified by their similarity to the measurement results for complex permeability and permittivity and coefficients of reflection and absorption for an epoxy composite with 80 mass% of CIP reported in [18]. The imaginary part of permeability increases to 2 at a frequency of 1 GHz and reaches a level of 2.3 within the 2–6 GHz range. The real part of permittivity of the composites varied from 9 to 16 depending on the type of micropowders, while the imaginary part varied from 0 to 2. The high absorbance of the composite with micropowder R10 (compared to the absorption characteristics of composites with R20 and R100F-2) is attributable to higher hysteresis and residual losses in R10. According to GOST 13610–79, they are $(3-5)\cdot 10^{-6}$ m/A and 0.15–0.25, respectively, being approximately 2 times higher than the corresponding losses in R20 and R100F-2.

We note in conclusion that the production of silicone composites with a high CIP concentration allows one to obtain materials with high absorption coefficients and complex permeability values and matched impedance. Despite the high CIP concentration (above 80 mass%), the samples were poor conductors of electric current. Electromagnetic losses increased with an increase in CIP concentration in the composite. The samples containing micropowder R10 provided the highest absorbance (10–60 %) at electromagnetic radiation frequencies of 0.1–6 GHz. The reflection coefficients increased monotonically from 5 to 20 % within the studied frequency range.

## Conflict of interest

The authors declare that they have no conflict of interest.

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