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Frequency and concentration dependences of the electrical properties of natural disordered carbon in the high-frequency region

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Abstract. Disordered sp^2 carbon of geological origin (shungite rocks of Karelia) with a set of various nanostructures (fullerene-like, graphene, ribbons) has promising technological properties. In this work, we studied the effect of nanosized carbon structures on the electrical properties. The impedance, active and reactance resistance, inductance of carbon-containing materials were measured in the frequency range from 50 kHz to 15 MHz. The inductive nature of the conductivity of shungite carbon was found. With increasing frequency, the resistance of shungite carbon increases. This nature of conductivity can be associated with the predominant effect on the electrical properties in shungites of nanosized ribbon structures.

Keywords: disordered sp^2 carbon, nanostructure, electrophysical characteristics

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Частотные и концентрационные зависимости электрических свойств природного неупорядоченного углерода в высокочастотной области

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Аннотация. Перспективными технологическими свойствами обладает разупорядоченный sp^2-углерод геологического происхождения (шунгитовые породы Карелии) с набором различных наноструктур (фуллереноподобная, графеновая, ленточная). В данной работе мы исследовали влияние наноразмерных углеродных структур на электрические свойства. Измерялись импеданс, активное и реактивное сопротивление, индуктивность углеродсодержащих в диапазоне частот от 50 кГц до 15 МГц. Обнаружен индуктивный характер электропроводности шунгитового углерода. С увеличением частоты сопротивление шунгитового углерода увеличивается. Такой характер проводимости может быть связан с преимущественным влиянием на электрические свойства в шунгитах наноразмерных ленточных структур.

Ключевые слова: разупорядоченный sp^2-углерод, наноструктура, электрофизические характеристики

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Introduction

In the shungite rocks of Karelia, disordered sp^2 carbon with a set of various nanostructures (fullerene-like, graphene, ribbon) was formed under geological conditions [1]. Promising technological properties of shungites (high electrical conductivity, reflection and absorption of microwave radiation, chemical stability and heat resistance) are associated with these nanostructures and porosity [2–9]. The carbon content in shungites varies from 2 to 97 at. %, and inclusions of minerals (mainly quartz) control some of their physical properties. In general, shungite is a natural carbon-mineral composite with alternating conductive (carbon) and non conductive (quartz) areas. Electrically conductive properties serve as the key both to the knowledge of the structural features of shungite carbon and to the expansion of promising technological applications. Previous studies of the frequency dependence of electrical properties on carbon content in the frequency range 100 Hz–200 kHz showed [10] that significant changes in electrical properties with increasing frequency start at frequencies of the first hundreds of kHz. In this regard, it is of interest to study the electrophysical characteristics of shungites and disordered carbon in a wider frequency range, primarily megahertz.

In this paper we present an analysis of the effect of nanosized carbon structures on the electrical properties based on modern structural studies of shungites and the frequency dependences of the total impedance, active and reactive resistances, phase angle, dielectric loss tangent, and specific conductivity of shungite samples.

Experimental

The measurement of the frequency dependence of complex resistance (impedance) is widely used to study the electrophysical properties of porous carbon materials. This paper presents the averaged results of measurements of the impedance and the phase angle, as well as the results of calculations of active and reactive resistances, the tangent of the dielectric loss angle, inductance and conductivity of shungite samples depending on the frequency and carbon content.

To study the conductive properties shungite samples with a carbon content of 5 to 97 at. % were used. The shungites can be conditionally divided into samples with high, medium and low carbon content. An E7-29 immittance meter (Minsk Scientific and Research Instrument-Making Institute, Belarus) was used to measure the impedance and phase angle in the frequency range from 50 kHz to 15 MHz.

The samples had the shape of a square tablet with side sizes from 7 to 10 mm and with thickness 1.5 mm. Measurements of inductance, impedance and phase angle were carried out in six separate areas for shungite with a carbon concentration below 50 at. %, and in 3–5 areas for shungites with a carbon concentration above 50 at. %. All measurements were carried out at room temperature.

Results

Figure 1 shows the dependence of the impedance modulus of shungite on the carbon content. The shungite impedance decreases with a carbon content increasing. The effect of the carbon content on the impedance Z is especially strong in the range of 5–35 at. %. At carbon content above 35 at. %, the decrease in the impedance value is much weaker.

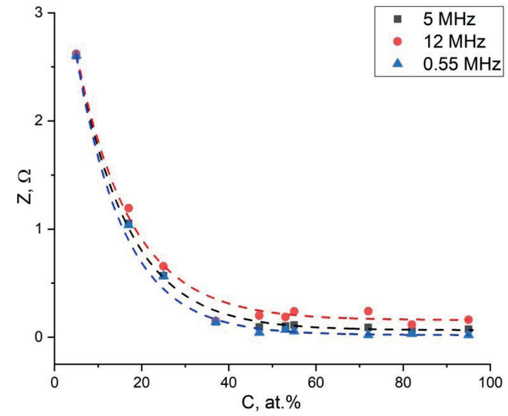

Fig. 1. Dependence of impedance on carbon content in shungites

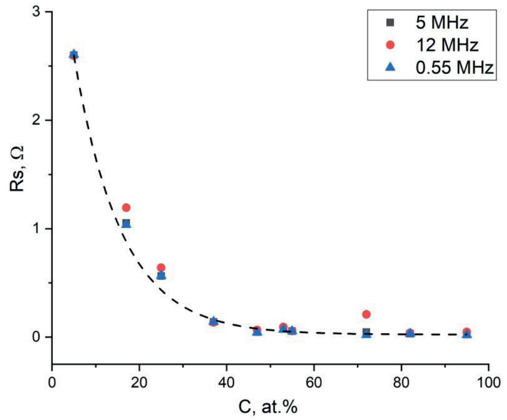

Fig. 2. Dependences of active resistance on carbon content

Phase angle θ and impedance Z are related to active R and reactance X by the following expressions:

$$R = \frac{|Z|}{\sqrt{\tan^2\theta + 1}} = |Z|\cos\theta, \tag{1}$$

$$X = \frac{|Z|\tan\theta}{\sqrt{\tan^2\theta + 1}} = |Z|\sin\theta. \tag{2}$$

Using these expressions, we calculated active (Fig. 2) and reactance resistances and conductivity (Fig. 3).

The largest contribution of inductance to reactance occurs in the range of 50 kHz–15 MHz. We estimated the value of inductance for shungite samples in the indicated range (Fig. 4). For all samples of shungite the inductance is approximately at the same level (2 ± 1) *nH*. Only for the sample with the lowest carbon content, the value of the inductance is much larger.

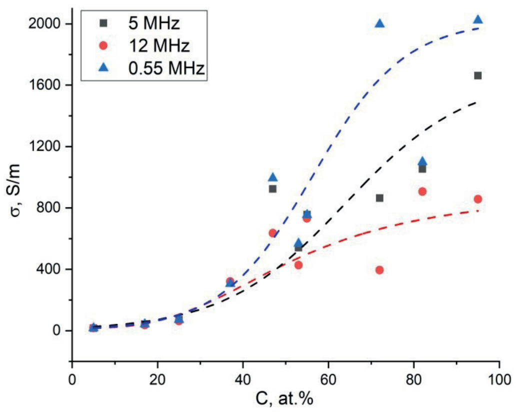

Fig. 3. Conductivity vs carbon content

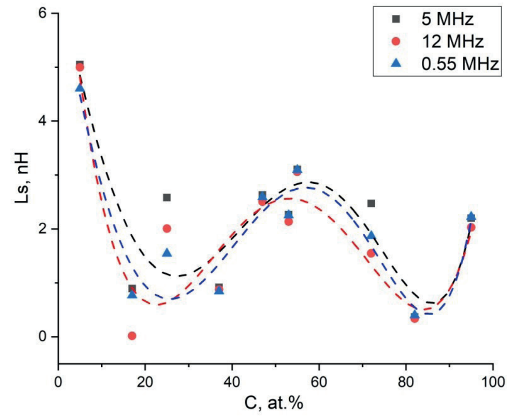

Fig. 4. Inductance vs carbon content

Discussion

For shungites the dependence of conductivity on frequency increases with increasing carbon content, and this dependence becomes significant at a carbon content of about 35%. The complex resistance (impedance Z) practically does not depend on the frequency. This is because active resistance dominates in shungites with a carbon content of up to 35%. The reactive part of the resistance is present, but its absolute values are small (0.1–0.4 Ω), even for samples with a low carbon content, where the reactance is greatest. At high values of active resistance for low-carbon samples (up to C = 35%), the contribution of reactance is very small, and there is no effect of reactance on the total conductivity. Starting from C = 35%, the value of active resistance drops sharply, and reactance begins to play a more significant role.

The increase in reactive resistance with frequency (Fig. 4) indicates the predominance of inductive resistance in shungites. The largest absolute value of the inductance is in samples with the lowest carbon content.

The inductive nature of conductivity at frequencies of 50 kHz–15 MHz can be associated with multilayer ribbon and fullerene-like structures of shungite carbon (Fig. 5), as well as with non-conductive inclusions up to several micrometers in size, which are covered with a thin film of ordered graphite carbon. Such structures can act as inductors or solenoids. The geometric parameters (the length and thickness of graphene-layer ribbons, the diameter of the turns) provide the predominant contribution of such structures to the inductive electrophysical properties in the measured frequency range.

The decrease in conductivity with increasing current frequency may be due to the routine mechanism of the increase in active resistance in the AC circuit. This mechanism consists in the uneven distribution of alternating current over the cross section of the conductor under the action of electromotive force (EMF) of self-induction. EMF is induced in the conductor by a magnetic field, which is created by the current passing through the conductor, and displaces the current to the surface of the conductor, reducing its useful cross section. If ribbon carbon structures with a thickness of 2–5 nm (Fig. 5), consisting of 5–15 graphene layers, are considered as key conductors in shungites, then such tapes correspond to the key paths of current propagation in shungites.

Fig. 5. Typical ribbon-graphene structure of shungite carbon

Conclusion

In the presented work, the electrophysical properties (impedance, active and reactive resistance, inductance) of carbon-containing natural composites (shungites) were measured at frequencies 50 kHz–15 MHz and calculated. Shungite samples with carbon content of 5–97 at. % were studied. These electrophysical properties were analyzed as frequency and concentration dependences.

A successively increasing dependence of the conductivity on the current frequency with an increase in the carbon content is found. The conductivity decreases with increasing current frequency. The complex resistance is practically independent of frequency due to the predominance of the active component.

The electrophysical properties can first be associated with ribbon carbon nanostructures in shungites, which create an inductive reactance and increase the active resistance in alternating current.

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