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Laser-Intercalation Processes in Carbon Material

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It is shown that diffraction lines corresponding to interlayer distances are shifted towards smaller angles. The double interlayer distance in this case increases by 0.005 nm compared to the initial graphite and amounts to 0.672 nm, which corresponds to the stage I intercalation mechanism. However, an increase in the half-width of the diffraction lines by 1.8 times compared to the initial graphite indicates the uneven penetration of the intercalant throughout the sample volume. In the diffractogram of the intercalated graphite irradiated by a laser, practically only diffraction reflections corresponding to the interlayer lines are observed.

Keywords: intercalation, sulfuric acid, carbon material, laser irradiation.

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Introduction

Known methods of intercalation, defined as the process of introducing ions, atoms, or molecules into the "guest" positions of a "host" material, are not without a number of drawbacks, which often prevent the synthesis of intercalates with the required properties. In particular, these methods do not allow for the uniform filling of "guest" positions throughout the sample volume; additionally, in some cases, their application requires a significant thermal load on the "host" material, which is often incompatible with the crystal lattice stability [1-2].

One possible way to eliminate these drawbacks in layered structures is to stimulate the intercalation process by irradiating a thin absorbing film of the "guest" component – deposited on one of the faces of the "host" material perpendicular to the layered crystal structure – with laser pulses. Irradiation was carried out both from the side of the deposited film and from the opposite side if the "host" material is transparent to laser radiation. In this case, as a result of the absorption of laser radiation by the "guest" component, it evaporates within 2-8 ms. Due to large concentration and temperature gradients, it is then introduced into the "guest" positions, which in layered crystals are located between atomic planes bound by weak van der Waals forces. Thus, by adjusting the thickness of the deposited film and the energy density of the laser radiation, the thermal load on the "host" material can be easily minimized, and the introduced "guest" component amount can be conveniently and effectively controlled by the number of laser pulses [3].

I. Experimental, discussion of the results

A characteristic feature of "host-guest" intercalate complexes is their ability to be effectively modified by external physical fields, in particular, by laser irradiation. The development of scientific research in this direction is of interest not only from a fundamental point of view but also from an applied one, as a technological approach to forming capacitor structures for alternating current circuits. One such complex is carbon material (graphite) intercalated with sulfuric acid, the laser irradiation of which significantly changes its structure and properties. It has been established that the preliminary intercalation of sulfuric acid into graphite leads to an almost fourfold increase in the lithium "guest" load. The measured specific capacity of the formed $Li_xC<H_2SO_4>$ co-intercalate reached a value close to 700 mAh/g, which is highly promising for its use as an anode material (Fig. 1). At the same time, the Nyquist plot, modeled by a parallel-connected R_{ct} and a constant phase element with a small phase deviation, is transformed by shifting to the right along the ReZ axis, with an increase in the complex impedance modulus by almost 100 times at a frequency of 10^5 Hz.

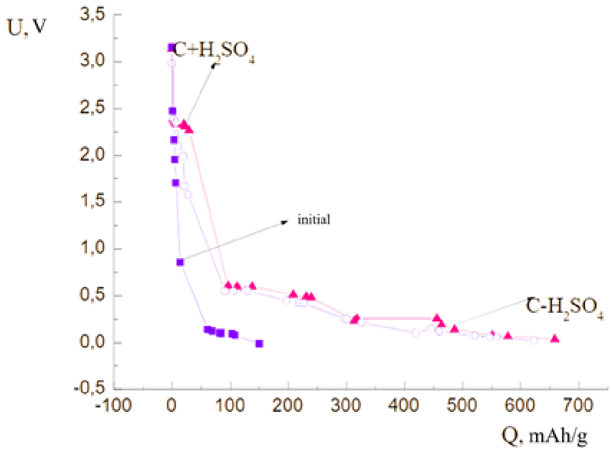


Fig. 1. Effect of H_2SO_4 on the discharge capacity of the complex during its lithium co-intercalation.

For the intercalation activation of graphite, cobalt fluoride (CoF_2) was used. The intercalation process was carried out at a temperature of 1200 K for 3 hours in an argon flow. The heating rate up to 700 K was 20 K/h, followed by a holding period of 20 hours. Heating to a temperature of 1200 K was performed at a rate of 60 K/h, and the cooling rate was 50 K/h. Deintercalation and drying were carried out according to the scheme described above for powdered talc. As can be seen from Fig. 2, we have a situation similar to intercalation-modified talc in terms of charge transfer between the liquid and solid phases – a drop in the charge transfer stage resistance. However, diffusion processes are less intensive, most likely due to the incomplete removal of cobalt fluoride during deintercalation. The studies were conducted on an automated X-ray diffractometer DRON-3 using CoK_α radiation. The recording was carried out in point-by-point registration mode: scanning step $\Lambda 2\theta = 0.1°$, exposure time $\tau = 3$ s with data output to a computer.

In the diffractogram of the intercalated finely dispersed graphite, no additional reflections were found compared to the initial graphite (Fig. 3). Lines from graphite and impurity phases are present. However, the diffraction lines corresponding to interlayer distances are shifted towards smaller angles. The double interlayer distance in this case increases by 0.005 nm compared to the initial graphite and is 0.672 nm (corresponds to the stage I intercalation mechanism). However, a 1.8-fold increase in the half-width of the diffraction lines compared to the initial graphite indicates the uneven penetration of the intercalant throughout the sample volume. In the diffractogram of laser-irradiated intercalated graphite, practically only diffraction reflections corresponding to interlayer lines are observed. Reflections from impurity phases are completely absent.

Laser irradiation was carried out using a YAG laser operating in Q-switched mode; the pulse repetition rate was 27-54 Hz.

The intensity of graphite reflections not related to interlayer distances is sharply reduced. The double interlayer distance decreases in this case to a value of 0.67 nm. At the same time, the half-width of the diffraction lines decreases, which is caused by an increase in the structural perfection of graphite crystallites. Thus, the laser irradiation of intercalated finely dispersed graphite leads to complete purification from impurity phases, a partial reduction in the intercalant content, and an increase in the structural perfection of graphite crystallites. It was experimentally established that a single laser irradiation of graphite-sulfuric acid intercalation compounds ($E = 1.2$ J/cm^2, pulse duration 15 ns) leads to a slight shift of the resonance peak of the capacity (which appeared after sulfuric acid intercalation) of the electrical double layer at the interface with the electrolyte into the low-frequency region, with a small increase in the resistance of the charge transfer stage of potassium intercalation. Doubling the energy of laser irradiation causes a strong shift of this peak into the high-frequency region (Fig. 4) with a simultaneous increase in its absolute value, which reaches a high value of ~ 300 mF/g in the kilohertz range.

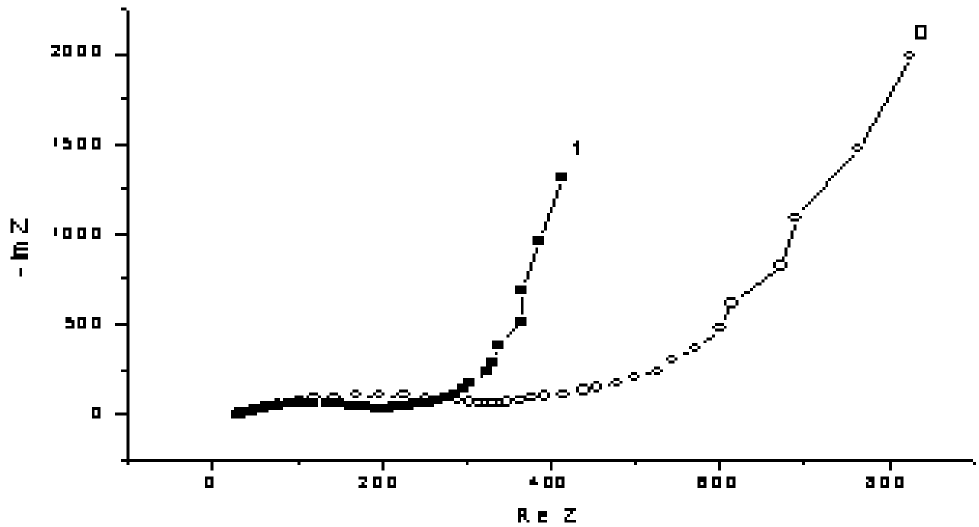
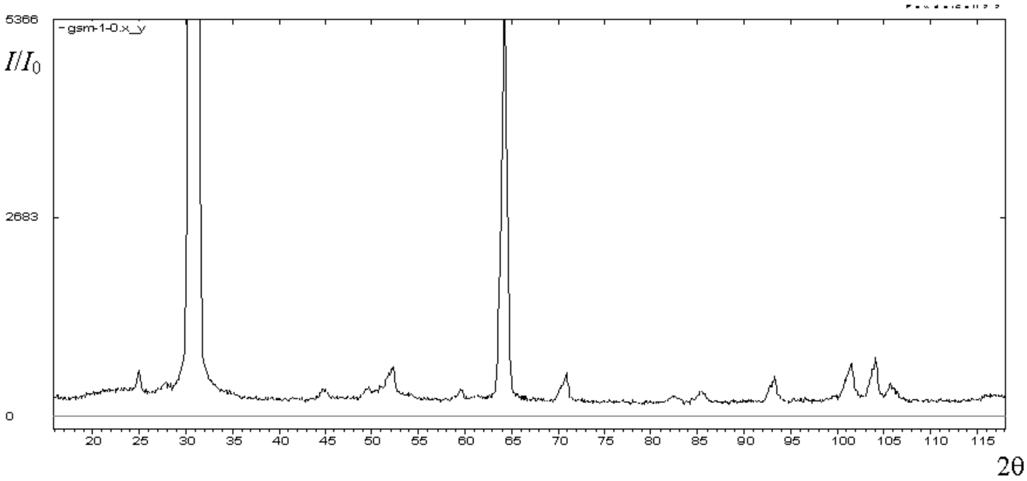
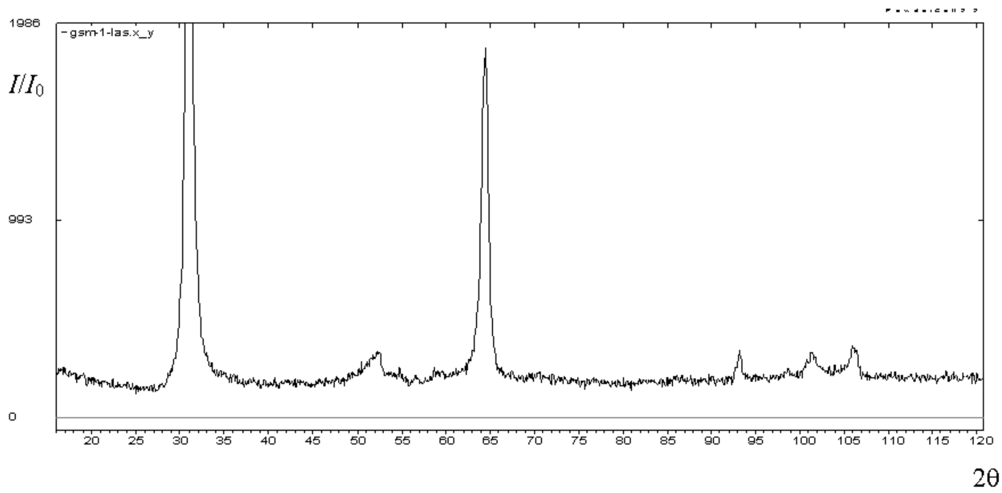


Fig. 2. Nyquist plots for the initial (0) and intercalation-modified (1) graphite.



a



b

Fig. 3. Diffractograms of the initial (a) and laser-irradiated (b) finely dispersed graphite.

Laser irradiation under the same conditions of the C<H_2SO_4> intercalate significantly reduces the charge transfer stage resistance during lithium co-intercalation and partially reverses the shift along the ReZ axis of the Nyquist plot caused by the introduction of sulfuric acid (Fig. 5).

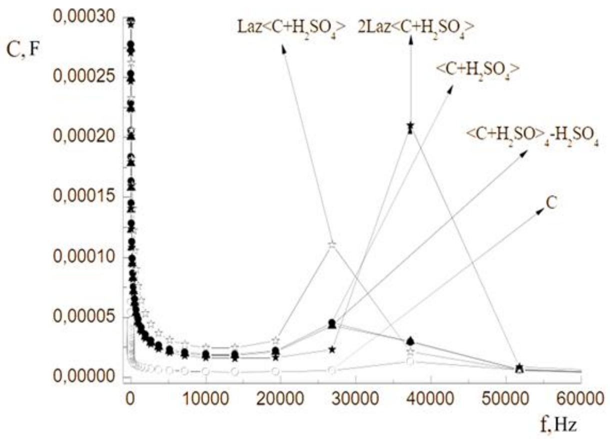


Fig. 4. The effect of laser irradiation on the electrical double layer capacity at the interface electrolyte – C<H_2SO_4> intercalate.

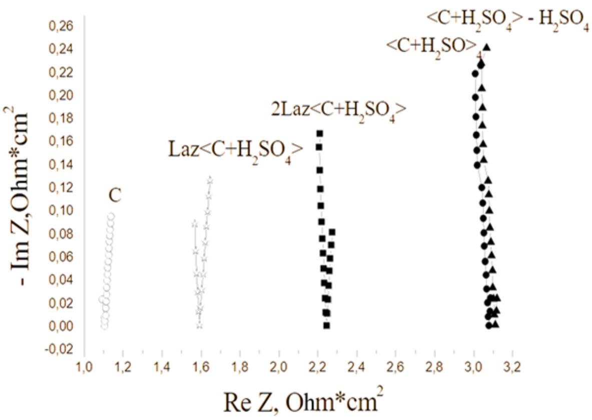


Fig. 5. The effect of sulfuric acid intercalation and laser irradiation of graphite on the shape of Nyquist plots.

Conclusions

1. It is shown that the intercalation of sulfuric acid into a carbon material (graphite) increases the lithium guest load (the amount of introduced lithium) by almost 4 times.

At the same time, the specific capacity of the obtained Li_xC<H_2SO_4> complex is 700 mAh/g.

2. According to the analysis of diffractograms from laser-irradiated complexes, there is a complete purification from impurity phases, a partial decrease in the H_2SO_4 content, and an increase in the structural perfection of the graphite.

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Лазерно-інтеркаляційні процеси в вуглецевому матеріалі

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Показано, що дифракційні лінії, що відповідають міжшаровим віддалям, зсунуті в сторону менших кутів. Подвійна міжшарова віддаль у цьому випадку зростає порівняно з вихідним графітом на 0,005 нм і становить 0,672 нм, що відповідає механізму інтеркалювання I ступеня. Однак, збільшення півширини дифракційних ліній у 1,8 рази в порівнянні з вихідним графітом свідчить про нерівномірність входження інтеркалянта по об’єму зразка. На дифрактограмі від інтеркальованого графіту, опроміненого лазером, простежуються практично лише дифракційні рефлекси, що відповідають міжшаровим лініям.

Ключові слова: інтеркаляція, сірчана кислота, вуглецевий матеріал, лазерне опромінення.