Investigation of the Process of Absorption of Microwave Radiation Generated by a Powerful Gyrotron in Powder Mixtures Al₂O₃/Pd

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Abstract. In this work, we investigated the energy input of high-power gyrotron microwave radiation into mixtures of Al₂O₃/Pd powders for various concentrations of the initial components. It was found that the absorption coefficient of microwave radiation as a result of the start of microdischarges (breakdowns) in the powder mixture depends on the Pd concentration in the initial mixture with aluminium oxide (Al₂O₃) in a complex way. It is shown that the addition of a catalyst, carborane (C₂H₂B₈), with a concentration equal to 10% of the mass of the initial Al₂O₃/Pd mixture increases the number of breakdown in the experimental session.

1. Introduction
In recent years, new methods of plasma-chemical synthesis of different micro- and nanoparticles based on the absorption of high-power microwave radiation by metal–dielectric mixtures are being rapidly developed [1]. The synthesized micro- and nanoparticles can be used for the manufacture of heterogeneous catalysts [2]. It is known that such catalysts of the platinum group (Pd / Pt) are used in the reactions of oxidation and dehydration of cycloalkanes [3]. In [4], a detailed study of the synthesis of microparticles based on mixtures of Ti + B, Mo + B and W + B powders was carried out. It was noted that for the formation of microwave microdischarges, which initiate plasma-chemical interactions between the particles of the mixture, it is important to match the parameters of the microwave pulse of the gyrotron, the particle size, and the mass ratio of the components in the initial metal–dielectric powder mixture. Changes in the absorption coefficient of microwave radiation in the metal–dielectric powder mixture can serve as a qualitative characteristic for analyzing the processes occurring in the plasma-chemical reactor [6].

Microwave radiation of gyrotrons can be used for producing nanoparticles of metal oxides by the evaporation–condensation method [6–8] and for ecological applications [9, 10]. In [11], the temperature dependence of microwave absorption in the gyrotron setup with a frequency of 263 GHz and maximum level power to 1 kW in powders of various metal oxides (Al₂O₃, ZnO, WO₃ and TiO₂) was studied by the calorimetric method. For Al₂O₃ powder increase of the absorption coefficient was observed with an increase in powder temperature. This fact plays an important role in our experiments because the absorbed microwave power of the gyrotron before the initiation of the breakdown and the development of the chemical reaction will be used for the heating of the metal–dielectric powder
mixture. Consequently, determining the absorption coefficient of the powder will allow us to control the synthesis process and predict the threshold of the microwave breakdown in the powder mixture from the microwave radiation power.

2. Experimental setup and measurement methods
The plasma chemical reactor is shown in Fig. 1. It includes an aluminum case with nozzle for connection to gas line 1, diagnostic window 2 and quartz input window 3. The reactor can be operated both at atmospheric pressure (in air and nitrogen) and in forevacuum. The initial metal–dielectric mixture of powders is placed on a quartz window 3.

![Figure 1. Photo of the plasma chemical reactor.](image1)

The schematic diagram of the experimental setup for studying plasma-chemical synthesis is shown in Fig. 2 [1]. The pulse of gyrotron 1 passes through the quasi-optical tract 2 and falls on the lower quartz plate of the plasma-chemical reactor 3, inside which a mixture of Al₂O₃ and metal Pd powders is placed. The range of the gyrotron power variation is from 100 to 400 kW, with pulse duration from 2 to 10 ms.

![Figure 2. Experimental diagram for calibrating microwave sensors for balance measurements.](image2)
To determine the absorption coefficient of microwave radiation of the gyrotron in the mixture of powders, a technique based on balance power measurements [12] was proposed in [4]. The power of gyrotron radiation absorbed in the powders can be calculated from the balance relation \( P_{\text{abs}} = P_{\text{in}} - P_{\text{ref}} - P_{\text{tr}} \), where \( P_{\text{in}} \) is the radiation power entering the plasma-chemical reactor, \( P_{\text{ref}} \) is the radiation power reflected from the parts of the quasi-optical tract and the elements of the plasma chemical reactor, \( P_{\text{tr}} \) is the radiation power transmitted through plasma-chemical reactor. The absorption coefficient is the \( P_{\text{abs}}/P_{\text{in}} \) ratio. For different Pd concentrations in the \( \text{Al}_2\text{O}_3 \) powder, the number of pulses in the series can differ because the series continues until the powders are exhausted, when after a certain number of microwave pulses the absorption coefficient decreases below 0.2. This value of the absorption coefficient was obtained experimentally based on observations of the breakdown threshold for the metal–dielectric powder mixtures in [1, 2].

To determine the radiation power, 3 standard D-407 sensors are used, two of which are located inside the quasi-optical coupler 3 (Fig. 2) and the third is located above the plasma-chemical reactor. All three sensors had to be calibrated to absolute power before experimenting. To calibrate the sensors, three modifications of the original experimental scheme were developed (Fig. 2). When calibrating the sensor for incoming radiation, the mirror that directed the radiation into the plasma-chemical reactor was rotated 90º, so that the radiation was directed into the volume of the flow water calorimeter with Teflon walls. Measurements of the water temperature in the calorimeter were conducted using two platinum thermal resistances. The reflected radiation sensor was calibrated using a mica plate with a reflection coefficient of 0.1, which replaced the plasma-chemical reactor. To calibrate the power sensor for power transmitted through the plasma-chemical reactor, at the beginning of the experimental campaign, a series of pulses were generated into an empty reactor. All sensors were calibrated with the same parameters of the radiation of the gyrotron: pulsed microwave power of 200 kW and pulse duration of 9.5 ms. The D-407 microwave sensor is a standard point detector of microwave power from several microwatts to several milliwatts. Since the experiments are conducted at relatively high values of microwave power (from 15 to 400 kW), a damping diaphragm is placed in front of the sensor, which substantially decreases the level of power absorbed in it, and thus, the correct choice of the diaphragm allows us to use the D-407 sensor in the linear regime with respect to microwave radiation power.

3. Results and discussion

Figure 3 shows the evolution of the absolute power of microwave radiation in a mixture of \( \text{Al}_2\text{O}_3/\text{Pd} \) powders with 5% palladium concentration. At a microwave power of 200 kW, no plasma-chemical reactions are initiated in the mixture of powders and the absorption coefficient is lower than 0.1 (Fig. 4). When the microwave radiation power is increased to 250 kW, initiation (excitation) of the plasma-chemical reactions is observed, which is accompanied by an increase in the absorption coefficient to 0.6. The microwave radiation power, at which the absorption coefficient above 0.2 is reached is determined as the threshold power of the initiation of the plasma-chemical reaction. In the experimental series under study, the number of pulses generated before the depletion of the powder mixture is 10. Fluctuations of the absolute power of the input microwave radiation are observed and the deviation from the nominal threshold power value in the calculation does not exceed 10%. This behavior of the absolute power value can have several reasons: first, the high probability of microwave radiation reflected from the plasma-chemical reactor entering the channel of the input microwave sensor, and second, the principle of operation of the gyrotron and the power supply of electrical pulses, which does not allow the output of constant microwave power during each pulse.

After depletion of the mixture of powders, a carborane-based catalyst was added to them, which is necessary to initiate the plasma-chemical reactions when the powder is depleted. The addition of the amount of carborane equal to 10% of the total mass of the mixture of \( \text{Al}_2\text{O}_3/\text{Pd} \) powders made it possible to extend the series by 2 pulses. After the depletion of the mixture of powders and carborane, the power of microwave radiation was increased to 300 kW, which made it possible to extend the series by a further 4 pulses.
**Figure 3.** Evolution of the absolute power of the incident (---), transmitted (-) and reflected (-) microwave radiation in the mixture of Al$_2$O$_3$/Pd powders with 5% of Pd in several consecutive pulses selected from a single experimental series.

**Figure 4.** Evolution of the absorption coefficient of microwave radiation power in a mixture of Al$_2$O$_3$ / Pd powders with 5% of Pd in several consecutive pulses selected from a single experimental series.
Table 1 presents the main results of calculating the absorption coefficients and the maximum absorbed radiation power of the gyrotron in the Al₂O₃/Pd mixture at various palladium concentrations. The absorption coefficient was calculated as the average of a series in which the deviation from two adjacent values did not exceed 20%.

**Table 1.** The results of calculating the absorption coefficient and the maximum absorbed power in a mixture of Al₂O₃/ Pd powders.

| Pd content, % | Carborane (C₂H₂B₈, 10% of total mass) | Absorption coefficient | Absorbed power, kW | Breakdown power threshold, kW | Number of pulses in the series |
|----------------|--------------------------------------|------------------------|-------------------|-----------------------------|-----------------------------|
| 1              | absent                               | 0.3820                 | 95.5              | 250                         | 5                           |
| 5              | absent                               | 0.8292                 | 207.3             | 250                         | 10                          |
| 10             | absent                               | 0.7824                 | 195.6             | 250                         | 6                           |
| 1              | added                                | 0.0853                 | 21.3              | 250                         | 3                           |
| 5              | added                                | 0.8211                 | 205.3             | 250                         | 2                           |
| 10             | added                                | 0.7957                 | 198.9             | 250                         | 2                           |

For a mixture of Al₂O₃ (99%) with Pd (1%), the lowest absorbed power coefficient is observed, which can be explained by the insufficient density of metal–dielectric contacts for the development of a chain plasma-chemical reaction. In this case, the addition of carborane decreases the absorption coefficient, which can be associated with the addition of one more dielectric to the mixture. When the palladium concentration in the mixture is increased to 5%, a significant increase in the absorption coefficient up to 0.8292 is observed, while the absolute value of the absorbed power is 207.3 kW. The addition of carborane (10% of total mass) also allows us to increase the number of pulses, while the absorption coefficient does not change. An increase in the palladium concentration up to 10% leads to a decrease in the absorption coefficient, which can be associated with an increase in the reflection of microwave radiation from the palladium powder.

4. Conclusions

The study of the absorption coefficient of microwave radiation in Al₂O₃/Pd powder mixtures performed in this work can serve as a qualitative justification for choosing the modes of synthesis of nano- and microparticles for other powders under gyrotron irradiation. The difference between the absorption coefficients for the three different concentrations of palladium should be used when choosing the correct ratios in mixtures of the powders under study for plasma-chemical synthesis. The effect of adding carborane on the development of plasma-chemical reactions leads to an increase in the number of pulses and the absorption coefficient in the case of a mixture of Al₂O₃/Pd powders.

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