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Nitrization of soil by corona discharge

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Experimental and theoretical studies of the nitrization possibility of turf-podzolic soil have been carried out. The experimental results obtained show that the electrical conductivity of the sample of the arable horizon of turf-podzolic soil increases with the time of exposure to plasma, the electrical conductivity of the soil paste after treatment for 2 hours is 285 $\mu\text{Sm/cm}$, after treatment for 4 hours is 317 $\mu\text{Sm/cm}$, with a reference value of the electrical conductivity of the sample of 115 $\mu\text{Sm/cm}$. The nitrate content in the aqueous extract from the soil shows an increase in 2 hours to 14.5 mg/l, and a further decrease in 4 hours to 12.8 mg/l, relative to the control of 9 mg/l. Calculations show an increase in plasma concentrations of nitrogen components that enter the soil. Their accumulated concentrations reach $5 \times 10^{21} \text{ cm}^{-3}$, which is an order of magnitude close to the experimental value of the accumulated nitrates during the experiment. The results confirm the idea of the possibility of using corona discharge for nitrization of poor soils.

Keywords: nitrization, turf-podzolic soil, electrical conductivity, nitrates, air, experiment, calculations.

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Introduction

The tasks of soil treatment by gas discharge appear relevant from the perspective of applications in agriculture. They are associated with the disinfection of air and grain, improvement of soil fertility, modification of its components, changes in their conductivity and chemical composition, and the generation of active particles within them [1–7]. Our study is devoted to these issues. Wherein, we investigate the question of saturating the soil with nitrogen components, i.e., whether the action of a corona discharge can lead to soil nitrification.

As it is known [2], nitrogen is the most important nutrient element for all plants: it is part of such important organic substances as proteins, nucleic acids, nucleoproteins, chlorophyll, alkaloids, phosphatides, etc. The nitrogen content in plants varies significantly depending on the

plant species, their age, soil and climatic conditions of crop cultivation, etc. Nitrogenous compounds available to plants are formed mainly from soil organic matter as a result of its decomposition. Soil nitrogen reserves are replenished mainly through the nitrogen-fixing activity of free-living and nodule microorganisms and its input from atmospheric precipitation. To some extent, the nitrogen reserve in the soil is replenished by nitrogen from atmospheric precipitation.

According to literature data [2], precipitation brings from 2 to 11 kg of nitrogen per hectare annually. The known sources of replenishing natural nitrogen reserves are of practical interest, but they supply only a part of the nitrogen that is removed with agricultural crop yields. Therefore, measures should be taken to increase soil fertility and, first of all, to replenish its nitrogen reserves.

Experiments with the ^{15}N isotope [2] have shown that nitrogen losses due to volatilization average 15 % of that applied, sometimes reaching 30 %. The greatest amount of nitrogen lost due to volatilization is in the form of ammonia (NH_3), molecular nitrogen (N_2), and nitrous oxide (N_2O). A significant part of gaseous nitrogen is lost from the soil due to the denitrification process [2]. This process of reducing soil nitrate nitrogen to free gaseous nitrogen (N_2) occurs as a result of the vital activity of soil organisms. The denitrification process proceeds through a series of intermediate stages [2]



Molecular nitrogen and nitrous oxide are the main gaseous products of biological denitrification, through the volatilization of which nitrogen losses from soils occur. The objective of this work was to elucidate the question of whether it is possible to carry out soil nitrification using a corona discharge plasma generated in the air near the soil surface. That is, whether it is possible to weaken the implementation of process (1).

Our research was conducted both experimentally using a negative corona discharge and theoretically based on a model of the discharge plasma. The following considerations guided these studies [1, 8]. The drift time of formed ions over a typical electrode needle length in the electric field of a corona discharge, on the order of 5×10^{-6} – 5×10^{-7} s in air, is much less than the typical soil treatment time, which reaches 1–4 hours. The active particles formed in the air plasma also move rapidly toward the soil surface under the action of diffusion. Thus, the characteristic diffusion time of particles formed in the plasma to the soil surface is in the range of 0.3–1 s, which is also less than the soil treatment time by the corona discharge plasma, which reaches 1–4 hours. Therefore, it can be deemed that the formation of active particles in the air by the corona discharge, which under our conditions influence processes in the soil, is an important result of the plasma action.

The influence of the humidity factor on the results of the corona discharge action is of undoubted interest, so in our calculations we used a humid air model, which differs from the model in [9]. This paper presents the results of model

studies to assess the influence of corona discharge plasma on the properties of the surface air layer near the soil surface at a real humidity level reaching 2 %, whose ionic components penetrate the soil.

Experiment

In the experiments, a corona discharge in air was used at voltages of 5–10 kV, with a current on the order of 100 μA , a description of which is presented in [7]. The discharge action time varied from 30 minutes to 4 hours. A negative corona was used as the test discharge in the experiment. The nitrate content was determined in a water extract at a soil-to-water ratio of 1:5. The products of chemical reactions in the soil pass into the solution. The nitrate content in the solution was measured by the potentiometric method using ion-selective electrodes on EXPERT-001 ionometer. The instrument is calibrated for a specific ion using standard solutions. KNO_3 is used for nitrates.

A sample from the plow horizon of an agrosoddy-podzolic light loamy soil on mantle loams underlain by moraine was used. Coordinates of the soil sample section X: 56.03867°, Y: 37.16581°, Z: 215.0 BS (Baltic System). Location: Chashnikovo, Solnechnogorsky District, Moscow region

Basic properties of samples subjected to corona discharge:

1. Sample moisture content – in an air-dry state, in a state of hygroscopic moisture of 2.81 % (0.281 g H_2O /g soil).
2. Acidity $\text{pH} = 5.53$ – 5.56 . Humus content 2.73 %.
3. Cation exchange capacity (CEC) – 14.25 mg-eq/100 g soil.
4. Physical clay content (particles < 0.01 mm) 26.8 %.
5. Specific surface area 51.0 m^2/g .
6. Surface charge density 2.4×10^{-3} mg/ m^2 .

Experimental Results

The experiment showed that the electrical conductivity of the sample from the plow horizon of sod-podzolic soil increases with the time of

plasma exposure. The electrical conductivity of the soil paste after treatment for 2 hours equals 285 $\mu\text{S}/\text{cm}$, after treatment for 4 hours – 317 $\mu\text{S}/\text{cm}$, with a control value of the sample's electrical conductivity being 115 $\mu\text{S}/\text{cm}$. The oxidation-reduction potential (ORP) of the control sod-podzolic soil water extract (without treatment) is 169–170 mV. The ORP of the water extract from the soil increases to +187 mV after 2 hours of treatment, and then after 4 hours of treatment it decreases to +180 mV. The ORP of distilled water is 147 mV+. Initial soil moisture content is 2.8 %, steam-saturated moisture is 6.0 %; after soil treatment for 2 h – moisture content is 5.4 %; after soil treatment for 4 h – moisture content is 2.9 %.

The nitrate content in the water extract from the soil shows an increase to 14.5 mg/l over 2 hours and a further decrease upon treatment for 4 hours to 12.8 mg/l, relative to the control of 9 mg/l. Thus, over 4 hours with an input power of 1 W, there was an increase in nitrate concentration by ~ 40 %, which is greater than the maximum nitrogen loss value of 30 % due to volatilization. At the same time, an improvement in soil quality occurred, as shown by the increase in ORP and electrical conductivity.

Model Calculations

This section presents the results of model studies to assess the influence of corona discharge plasma on the properties of the surface air layer near the soil surface. Model calculations were performed using the Kinet program, with verification in the Maple 2024 software, employing the Rosenbrock method. We were primarily interested in N_xO_y compounds and their yield from reactions, since upon interaction with water they form HNO_3 and, according to calculations in [10], are the main source NO_3^- . Based on the analysis of reaction rate constants [8–12], 150 chemical reactions and 34 components involved in the transformations were included in the model, such as: e, O, O_2 , O_3 , O^- , O_2^- , O_3^- , O_4^- , O^+ , O_2^+ , O_4^+ , N, N_2 , N_2^+ , N_2O , NO, NO_2 , NO_3 , N_2O_5 , NO^+ , NO^- , NO_2^- , NO_3^- , NO_2^+ , H_2O , H_2O^+ , H_3O^+ , H_2 , H, H^- , OH, OH^- , HO_2 , HNO_2 , HNO_3 . Calculations were performed

for values of the reduced electric field: 150 Td and humidity of 2 % (in this case referring to the ratio of the concentration of water molecules in the air to the total concentration of components in the air). For humid air, the rate constant of dissociative attachment in the field at 150 Td was taken equal to $9.4 \times 10^{-10} \text{ cm}^3/\text{s}$, the rate constant of oxygen molecule dissociation in such a field was taken equal to $1.9 \times 10^{-8} \text{ cm}^3/\text{s}$, and the ionization rate constant was taken from [8].

In the case of an electric field strength of 150 Td, the concentration of O_3 increased to $3 \times 10^{18} \text{ cm}^{-3}$, OH (is consumed rapidly, its concentration drops over 6 seconds from $8 \times 10^{16} \text{ cm}^{-3}$ to $2 \times 10^0 \text{ cm}^{-3}$ (i.e., practically almost to zero), $\text{N}_2\text{O} \approx 2 \times 10^{14} \text{ cm}^{-3}$, $\text{NO} \approx 4.7 \times 10^{10} \text{ cm}^{-3}$, $\text{NO}_2 \approx 1.7 \times 10^{12} \text{ cm}^{-3}$, $\text{NO}_3 \approx 6 \times 10^{14} \text{ cm}^{-3}$, $\text{N}_2\text{O}_5 \approx 1.3 \times 10^{17} \text{ cm}^{-3}$, HNO_3 decreases to $4 \times 10^{10} \text{ cm}^{-3}$ considering the reverse reaction with HNO_2 , whose increase reaches a value of $3.2 \times 10^{17} \text{ cm}^{-3}$. This is seen from Fig. 1. This figure shows the evolution of the concentrations of the components HNO_3 and HNO_2 . Evidently, within the framework of the presented model, the components HNO_2 remain in the system, which participate in enriching the soil with nitrogen components as a result of the sequence of chemical reactions.

With an increase in air humidity, the electron concentration increased by up to two orders of magnitude, which in the model leads to an acceleration in obtaining components; for example, for ozone at a humidity of 1 %, the ozone O_3 concentration rises to a value of $3 \times 10^{18} \text{ cm}^{-3}$ in 50 μs , and in the absence of moisture in the same time. The maximum achieved values of the components differed, especially for nitrogen components.

Fig. 2 presents the calculation results for the concentration of negative ions at $E/N = 150$ Td in air at 2% H_2O . It can be seen that at long times, the ion concentrations reach steady-state values, with the maximum increase observed for the NO^- ion.

Fig. 3 presents the concentrations of components involving nitrogen and ozone molecules depending on different concentrations of H_2O in the air at $E/N = 150$ Td. It can be seen that the concentration of N_2O_5 is the main one among the nitrogen-containing components at all humidity values considered.

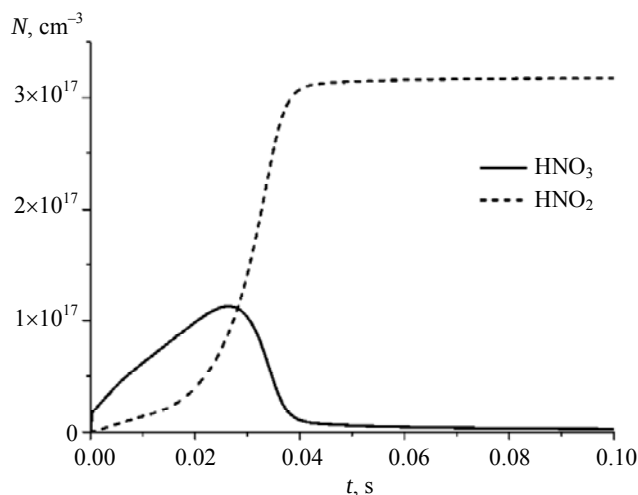


Fig. 1. Evolution of HNO_3 and HNO_2 (cm^{-3}) under discharge action at $E/N = 150$ Td in air at 2 % H_2O

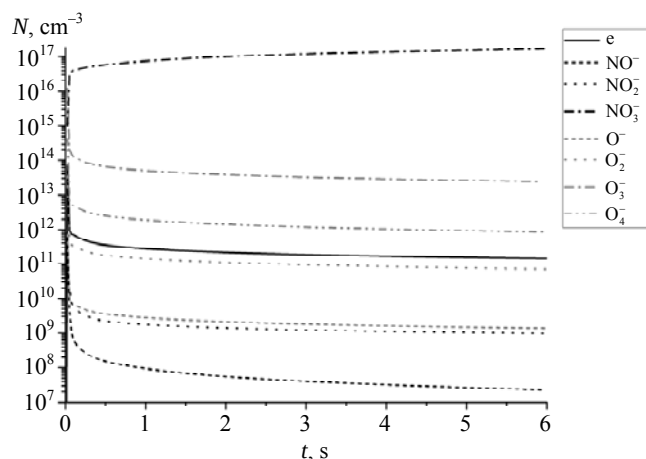


Fig. 2. Evolution of negative ion concentrations in air under discharge action at $E/N = 150$ Td in air at 2 % H_2O

Comparison with the models of Kossyi et al. [9] and Sekimoto et al. [10] confirms the correctness of the relative concentrations of the components and their dependence on the electric field; however, the absolute values of the concentrations of HNO_3 and N_2O_5 require further refinement within the framework of developing the theoretical model.

The model did not consider the results of work [12] on the role of positive nitrogen ions near the electrodes, nor the fact that particles composing the soil can enter the plasma. The obtained results allow a comparison of experimental and theoretical data.

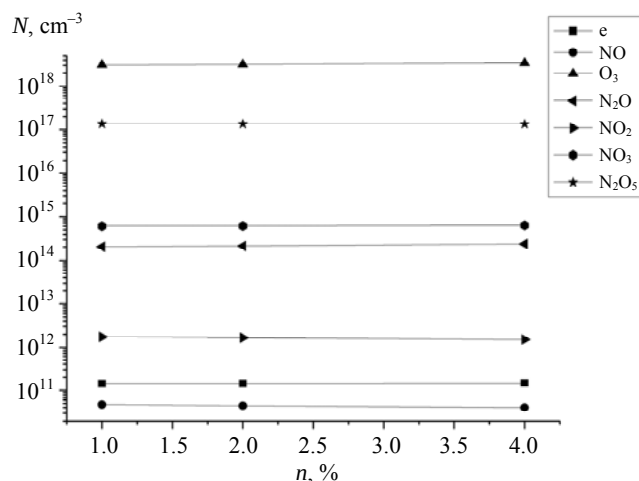


Fig. 3. Concentrations of neutrals for different concentrations of H_2O in air ($n = 1\%$, 2% , and 4%). The calculation times for humid air were $600\ \mu\text{s}$, and for dry air – up to $100\ \text{s}$, $E/N = 150$ Td

Measurements showed that the average nitrate content in the water extract from the soil over 2 and 4 hours increases by $5\ \text{mg/l}$ relative to the control, or $5 \times 10^{-4}\ \text{g/cm}^3$. We will consider HNO_2 as the reference component. From Fig. 1, the production rate of HNO_2 can be estimated as $Q = 10^{19}\ \text{cm}^{-3} \cdot \text{s}^{-1}$. This yields that over 4 hours, $2.4 \times 10^{21}\ \text{cm}^{-3}$ of HNO_2 will be produced in the volume, and a similar order of magnitude concentration of N_2O_5 will be produced, i.e., both nitrate production mechanisms together will yield a quantity on the order of $\sim 5 \times 10^{21}\ \text{cm}^{-3}$, which is of the same order of magnitude as the experimental value of nitrates produced. We see that efficient production of nitrates occurs.

Conclusion

The obtained experimental results show that the electrical conductivity of the sample from the plow horizon of sod-podzolic soil increases with the time of plasma exposure. The electrical conductivity of the soil paste after treatment for 2 hours equals $285\ \mu\text{S/cm}$, after treatment for 4 hours – $317\ \mu\text{S/cm}$, with a control value of the sample's electrical conductivity being $115\ \mu\text{S/cm}$. The oxidation-reduction potential of the water extract from the soil increases to $+187\ \text{mV}$ after 2 hours of treatment, and then after 4 hours of

treatment it decreases to +180 mV. This shows that an improvement in soil quality occurs. The nitrate content in the water extract from the soil shows an increase to 14.5 mg/l over 2 hours and a further decrease upon treatment for 4 hours to 12.8 mg/l, relative to the control of 9 mg/l. This latter result shows that the action of a low-power corona discharge leads to a positive effect on the properties of the surface soil layers. In this case, the nitrate content increases by an average of 50 %. The performed calculations show an

increase in the concentration of nitrogen components in the plasma, which enter the soil. Their produced concentrations reach $5 \times 10^{21} \text{ cm}^{-3}$, which is of the same order of magnitude as the experimental value of nitrates produced during the experiment. The obtained results confirm the idea of the possibility of using a corona discharge for the nitrification of poor soils. Therefore, the use of a corona discharge can be recommended as a means for soil activation in places of risky human activity.

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