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Ecological aspects of sorbents use to improve the efficiency of bioremediation on oil-contaminated lands

Aspectos ecológicos del uso de sorbentes para mejorar la eficiencia de bioremediación de suelos contaminados por petróleo

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Abstract

Nowadays, polluted soils are one of the primary environmental problems. Due to a large number of oil-contaminated lands, new methods are being developed more and more actively, as well as a combination of various existing methods for disposing of oil spills. In this paper, bioremediation with the use of adsorption is considered to reduce toxicity and accelerate the processes of microbial destruction of hydrocarbons. There is a large selection of sorbents based on carbon, mineral, and natural materials. The effectiveness of a particular sorbent will vary depending on the application region, weather and climate conditions, soil type, and the type and properties of the oil. In this work, peat was tested as an organic sorbent (S1), vermiculite as a mineral sorbent (S2), and sorbent based on carbon (S3). The sorbents were added to the soils contaminated by oil, evaluating the pH, humidity, and phytotoxicity of the soils. The results showed that the soils treated with sorbents maintained the neutral pH, increased the humidity, and decreased the phytotoxicity of the soil. The sorbent efficiency was S3> S2> S1.

Keywords: Environment, Bioremediation, Oil spill, Sorbent, Contaminated soil, Biodestruction, Petroleum Industry.

Resumen

Hoy, los suelos contaminados son uno de los principales problemas ambientales. Debido a la gran cantidad de tierras contaminadas con petróleo, se están desarrollando nuevos métodos más activamente, así como una combinación de diversos métodos existentes para la eliminación de derrames de petróleo. En este documento, se considera que la biorremediación con el uso de adsorción, reduce la toxicidad y acelera los procesos de destrucción microbiana de hidrocarburos. Hay una gran selección de sorbentes a base de carbono, minerales y materiales naturales. La efectividad de un sorbente en particular variará dependiendo de la región de aplicación, las condiciones climáticas y climáticas, el tipo de suelo y el tipo y propiedades del petróleo. En este trabajo, una turba fue probada como sorbente orgánico (S1), una vermiculita como sorbente mineral (S2) y un sorbente a base de carbono (S3). Los sorbentes fueron agregaron a los suelos contaminados por aceite evaluando el pH, la humedad y la fitotoxicidad de los suelos. Los resultados mostraron que los suelos tratados con sorbentes mantuvieron el pH neutro, aumentaron la humedad y disminuyeron la fitotoxicidad del suelo. La eficiencia del sorbente fue S3> S2> S1.

Palabras clave: *Medio ambiente, biorremediación, derrame de petróleo, sorbente, suelo contaminado, biodestrucción, industria petrolera.*

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Introducción

Wherever oil is extracted, transported, stored, and used, there is a risk of leakage. Spilled oil or petroleum products have a sharp unpleasant taste and smell, negatively affect all spheres of life, cause serious damage to the environment. Spilled oil eventually introduces toxic components into the human food chain and affects our health. Therefore, spills of oil and petroleum products cause huge environmental problems if the territory is not reclaimed as a matter of urgency. Today, the soil is subjected to intensive anthropogenic impact (Zinnatshina, Strijakova, Danshina, & Vasilyeva, 2018).

Pollution by oil and petroleum products has an extremely negative impact on the soil, reducing the quality and productivity of the land. Contaminated soil becomes toxic to plants, animals, and humans, and in some cases can lead to serious poisoning and death.

As a result of environmental catastrophes associated with accidents in fields where mineral extraction occurs, soil pores are clogged, which makes it hydrophobic, and usually leads to low water retention capacity. (Ahn et all., 2005)

Spills of oil and petroleum products are possible during the direct production, transportation, processing, and use of hydrocarbons. After entering the environment, oil or petroleum products negatively affect ecosystems, while significantly changing the pH level and the degree of aeration of the soil.

The need to eliminate the consequences of emergency spills is one of the primary tasks of the environment, and in this regard, methods for cleaning the soil from oil pollution are constantly being developed and refined. In this paper, we will consider the main factors that affect the rate of bioremediation of oil-contaminated soils, as well as the principles of using sorbents to speed up cleaning processes. (Akovetsky, Mikhedova, 2020).

Earlier, our laboratory developed a method for sorption bioremediation of oil-contaminated soils based on the use of various natural sorbents to accelerate the bioremediation process using the example of loamy gray forest soils.

The goal of our research is to develop a method of sorption bioremediation that can be used for the reclamation of oil-contaminated sandy soils found in the conditions of Western Siberia of the Russian Federation.

Theoretical framework

Bioremediation

Bioremediation is one of the most promising methods for cleaning oil-contaminated soils. It is based on the ability of microorganisms and other living organisms to decompose and utilize various hydrocarbons. In the natural environment, there is no waste, and biodegradation occurs continuously and everywhere; any organic substance can supply the entire complex of microorganisms with nutrients and eventually mineralize organic substances to form carbon dioxide and water.

Today, about 70 genera of bacteria and mucoromycetes are known, as well as some cyanobacteria and algae that can break down petroleum hydrocarbons. In nature, microorganisms exist in consortia, associations with each other and other organisms, showing their inherent functional activity.

The rate of degradation of hydrocarbons depends on the composition of the microbial population, the type, and the level of contamination. Although most soils usually contain native microorganisms that can decompose oil hydrocarbons, however, to improve the efficiency of bioremediation of oil-contaminated soils, it is also proposed to introduce biologics based on microorganisms that can use oil hydrocarbons as growth substrates.

For many years, biologics containing monocultures or consortia of microorganisms have been used to clean oil-contaminated soils. Some authors believe that the most promising is the use of adapted autochthonous microflora isolated from the soil of this polluted area by the method of accumulative crops.

The degree of degradation of organic compounds also depends on their structure. Simpler aliphatic and monoaromatic substances decompose relatively easily, while more complex structures require more effort to destroy them and can persist in the soil for a long time.

To overcome this problem, surfactants are used that are added to contaminated soils to increase the availability of high molecular weight hydrocarbons. (Akovetsky et al., 2020).

To provide the main biophilic elements for oil-destroying microorganisms, it is necessary to apply nitrogen, phosphorus, and potassium in the form of mineral or organic fertilizers. Another way to improve the efficiency of biodegradation of hydrocarbons can be to maintain optimal humidity and increase the degree of soil aeration by mixing and applying loosening additives in the form of wood chips, sawdust, or a waste of plant origin. (Mohan et all., 2006)

The rate of bioremediation of oil-contaminated soils depends on some factors, including the type of soil, water-salt regime, geographical location of the contaminated site, etc. The colder the climate, the lower the soil's ability to self-clean. Since a significant part of the oil fields in Russia is in the Arctic zone, it is necessary to develop methods for the rehabilitation of oil-contaminated soils for territories with a cold climate.

The relative ability of soils to self-purify from petroleum hydrocarbons is largely determined by their thermal regime. In regions where cold climate prevails, the bioremediation process must be intensified. However, despite the inestimable advantages of bioremediation, if there is a significant oil or petroleum product spill, the level of soil toxicity may be too high for active bioremediation. A method of sorption bioremediation based on the use of a wide range of sorbents was developed for cleaning highly polluted soils (Vasilyeva et al., 2019; Zinnatshina et al., 2018).

Currently, the use of sorbents is extremely important to prevent or reduce pollution and its environmental consequences. About 200 types of sorbents are used in the world to eliminate oil spills. Sorption techniques, as the name implies, utilize sorbents. This type of material can separate certain substances from other liquids or gases. Their useful characteristic comes from their porous nature and binding molecules to their solid surface. Their versatile capability in cleaning purposes has made them an attractive option for the petroleum industry to remove spills from the environment. Theory, this technology can be useful for any kind of surface, from coasts to roads or even contaminated vapors. What makes this method even more appealing is its cost-effectivity, besides safety and being user-friendly. Another ecological benefit of the Sorbent method is preventing further spills by making a barrier as a protective embankment. When they are no longer needed, these materials can be recycled, disposed of, or restored.

This category of materials, utilized in oil spill cleaning, origins consist of a few groups: inorganic mineral; natural organic; carbonaceous, and synthetic-organic (synthetic polymers) (Artyukh, Mazur, Ukraintseva, & Kostyuk, 2014).

Mineral sorbents are a large group; compared to other sorbents. They present many advantageous features such as chemical stability (especially non-combustibility), and of course, being inexpensive and easily available. Belonging to the sinking sorbent class, they have high density and a well-grained structure, in both natural and processed origins. This enables them to become suitable for absorbing floating oil.

Mineral sorbents could count as universal ones, mostly natural raw materials in form of powder or granule. They can be as small as nanometer-sized or up to three mm particles. Their chemical stability even extends to resistance to acid/base or even fire. When eliminating a ground oil spill, they are applied mechanically (using a brush) and distributed over the surface, and after the oil is absorbed, they are collected and sent for processing. Inorganic types of sorbents include various types of clay, loose diatomite rocks, various types of zeolites, tuffs, etc. The most popular inorganic sorbents are clays and diatomite since their cost is low and they can be produced in large volumes. Sand is also used for small spills, but the sand has a low oil capacity (from 70 to 150%) and is not able to sorb petroleum and gasoline fractions. The use of mineral materials is not possible on water surfaces due to the high density. Also, there are problems with their disposal. Usually, mineral sorbents are cleaned from the absorbed oil by solvent extraction, washing with water with Surfactants, or by burning. (Mikhedova, Abashina, 2020).

Like other types of sorbents, the Natural Organic group includes materials with high cost-benefit and nature-friendly, hence the name. Their biodegradability and ease of disposal with incineration are truly advantageous. Although, their low weight and density make them unsuitable for fire-hazardous conditions. The best members of this category are those which after some treatments (such as heat) gain superior absorption ability to oil. To name a few Natural Organic adsorbents useful for soil cleaning one could mention simple day-to-day materials such as linen, cotton, sawdust, wool, moss, leaf, or even paper cellulose. (Antizar-Ladislao et all., 2004)

Wool is an effective sorbent: 1 kg of wool can absorb up to 8-10 kg of oil or petroleum products, but the disadvantage of this sorbent is that after several squeezes, it is saturated with a bituminous fraction and its use becomes impossible. Another disadvantage is the high cost and complexity of storage (you need special conditions, protection from rodents, and insects). Wood sawdust is an effective sorbent, it absorbs hydrocarbons well and quickly, but it is hydrophilic, so when preparing sawdust as a sorbent, it is impregnated with water-repellent compounds (for example, fatty acids). Natural organic sorbents are becoming a suitable choice for cleaning up oil spills due to their availability, environmental friendliness, and low cost.

Synthetic polymers consist of polypropylene, polyethylene, polyacrylate, polystyrene, and polyurethane. They are normally components of manufacturing materials dealing with hazardous liquids, such as hoses. They are hydrophobic, with low bulk density and high sorption ability for oil. They have a wide variety of sorption capacities respective to the material to adsorb, from less than 10 g/g to dozens of h/g, or even one hundred. (Elssaidi et all., 2012)

Carbon sorbents are one of the most widely used in practice. This type of sorbent is obtained from carbon raw materials, such as peat, anthracite, wood, stone, or brown coal. There are many types of sorbents of this type, but the most popular are activated carbon (AC) and biochar. Due to their porous structure, they absorb fluids well. There are three main forms of particles: granulated (granules can be cylindrical or spherical), crushed (have a grainy irregular shape), in the form of powder (very small dust-like particles).

In the case of petroleum contamination, sorption can occur by adsorption. Oil can penetrate the porosities of the "absorbent" materials. While "adsorbents" only pull the oil to their surface but do not allow its penetration. The latter mechanism is a promising process and cost-effective method for reducing environmental problems. For the sorbent to be effective when used, it must have the following characteristics:

- High oil capacity and hydrophobicity.
- The buoyancy index after oil absorption is important.
- Ability to remove oil from the sorbent.
- The recyclability.

Often, when collecting oil or petroleum products, a combination of mechanical methods of collection and sorbents is used. Sorbents can absorb oil and do not allow the formation of an emulsion.

In recent years, research has been actively conducted to improve the environmental health and productivity of soils using various types of sorbents, including composite materials with microbiological and trace element additives.

Carbon materials and composites with some mineral and plant materials are promising for use as a carrier for microbial cells. Such carrier sorbents have high chemical resistance, mechanical strength, ion exchange properties, sufficient permeability to water and other substrates, biocompatibility, and manufacturability. The distinctive properties of the main carrier sorbents are that they were based on cheap and affordable pyrolysis products of plant waste from agriculture and woodworking, as well as enriched with trace elements zeolites and some other examples of natural aluminosilicates and oxides. (Gaur et all., 2018)

To obtain a sorption bio complex, which in its way is a catalyst in biotechnological processes, a careful study of the influence of the surface nature of the sorbent-carrier of microorganisms is required:

A. on the nature of sorption of the microbiological component.
B. For the preservation of its catalytic activity.

This allows us to predict the future use of this material. For the sorption bio complex to be competitive in natural conditions, the target carrier of the sorbent and the bacterial component must be selected properly. As for the bacterial component, in nature, bacteria are extremely numerous and have a flexible metabolism that allows them to live in any biosphere.

Whether xenobiotic destructive strains, after use, will survive in the soil and maintain their destructive activity in natural conditions are a question that has yet to be answered. In nature, micro-organisms that are destructors of various ecotoxicants are widely distributed. The population composition of their microflora changes in soils that have been repeatedly exposed to pollutants. (Antizar-Ladislao et all., 2005)

These conditions make the idea of bioremediation by utilizing nature-inspired methods, attractive for rectifying the problems caused by oil spills in the environment.

Although further field research is demanded to find the most cost-effective method for each climate. Furthermore, economical support would facilitate ecologic-friendly projects to become more applicable in everyday industrial efforts. And ideally applying remedial procedures as standards to be followed by oil companies in near future. This would solve many problems right after they occur before they lead to serious damages to the environment.

Methodology

1. Phytotesting was carried out by the express method developed by us - on the germination of white/creeping clover (Trifolium repens)
2. The soil in the vessels was sown with mustard seed (Sinapis).
3. The value of the water pH of the soil was determined in the water extract (1:2.5) using a pH meter
4. The humidity of the samples was determined by the thermostatic-weight method

Table 1. Experimental scheme.

	CC	C1	C2	S1	S2	S3
Soil (kg)	11	11	11	11	11	11
Oil (%)	-	6.5	6.5	6.5	6.5	6.5
Biological product (Microback)	-	+	-	+	+	+
Dolomite flour	+	+	+	+	+	+
Mineral fertilizers	+	+	+	+	+	+

For the experiment, sandy soil was taken from the territory of an oil field in the Khanty-Mansi autonomous region, Western Siberia, Russian Federation. Clean soil was polluted by 6.5% of oil. The oil is taken from a low-density characteristic of the region of production.

For the micro-field experiment, clean soil was taken, and then it was contaminated with crude oil. After 2 days, sorbents of 3 classes were added.

CC - clean control.
C1 - control without sorbent.
C2 - control without sorbent and biological product.
S1 - organic sorbent, peat.
S2 - mineral sorbent, vermiculite.
S3 - is a composite based on a carbon sorbent.

During the experiment, ph, humidity, and phytotoxicity were monitored in the samples. After measuring these indicators, depending on the results, a decision was made to add mineral fertilizers to the samples, as well as dolomite flour. By applying mineral fertilizers, we not only introduce useful substances into the soil, and activate the applied bacterial preparation, but also, we activate the native microflora located in the soil, thereby significantly accelerating the process of microbial degradation of hydrocarbons. Samples with soil were mixed and moistened. Mineral fertilizers are necessary for the work of microorganisms, under favorable climatic conditions, the number of microorganisms and their activity after fertilizing the soil significantly increases, and therefore the destruction of petroleum hydrocarbons occurs. With a lack of mineral fertilizers in the soil, there is a high probability of stopping microbial activity, and, accordingly, microbial oil destruction.

Results and discussion

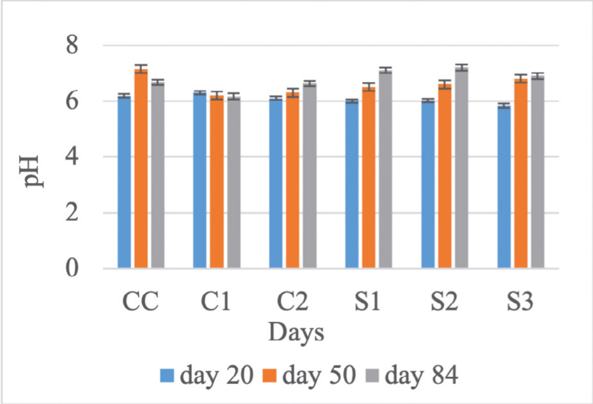

Figure 1. Effect of sorbents to the samples per day on pH.

To maintain the active process of oil degradation, the use of sorption bioremediation is effective. For this process, it is necessary to constantly monitor the acid-base balance of the samples. Graph 1 shows the results of the pH analysis of samples. The graph shows that over time, the pH is approaching the neutral value of the indicator. Sorption bioremediation contributes most effectively to this process. The difference between samples with and without sorbent is visible. On day 84, the pH balance of the C2 sample was significantly lower than that of the S1, S2, and S3 samples. Monitoring of the samples demonstrated the effect of the mineral sorbent on the pH value. For organic class sorbents, a more detailed observation is required, due to the initially complex sorption characteristics.

From Fig. 1 it can be seen that by 84 days, the pH value in samples S1, S2, and S3 came to a normal (neutral) value. In sample S1 on day 20, the pH value was 6, then after 30 days-6.5, and on day 84 it became 7.1. in samples S2 and S3, the pH values increased from 6.02 and 5.84 to 7.2 and 6.9, respectively. While in the control sample C1 (without sorbent), the pH values decreased from 6.3 (20 days), 6.2(50 days) to 6.17 (84 days). And in the C2 sample (without consent and biological products), the values increased slightly, from 6.1 (20 days), 6.3 (50 days) to 6.63 (84 days). In the CC (clean control) sample, the values varied from 6.19 (20 days), 7.15 (50 days) to 6.68 (84 days).

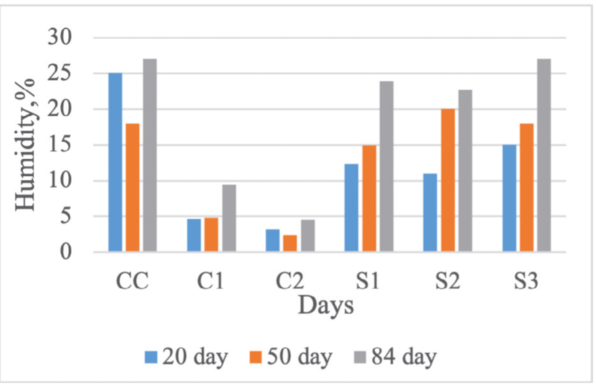

Figure 2. Influence of sorbents on the samples per day on humidity percentage.

When it enters the soil, the oil envelops the soil conglomerates, making it hydrophobic. As a result, oil seeps into the deeper layers of the soil, as well as groundwater. Graph 2 shows the beneficial effect of sorbents on increasing soil hydrophilicity in samples S1, S2, S3, and CC, while C1 and C2 remain hydrophobic despite the passage of time.

Figure 2 shows the effect of the presence of sorbents on the moisture level in the samples. In the CC sample, the humidity varied from 25% (day 20), 18% (day 50), and 27% (day 84). In the control sample C1 (without sorbent), the humidity values varied from 4.64% (day 20), 4.86% (day 50), and 9.4% (day 84). While in the sample C2 humidity is even lower than in C1 3.2% (day 20), 2.44% (day 50), 4.5% (day 84). At the same time, the humidity in the samples containing the sorbent S1, S2, S3 is noticeably higher. For example, in sample S1, humidity increased from 12.3% (day 20), 14.96% (day 50) to 23.95% (day 84). In the S2 sample, the humidity increased from 11% (day 20), 20% (day 50) to 22.71% (day 84). In the S3 sample, the humidity increased from 15% (day 20), 18% (day 50) to 27% (day 84)

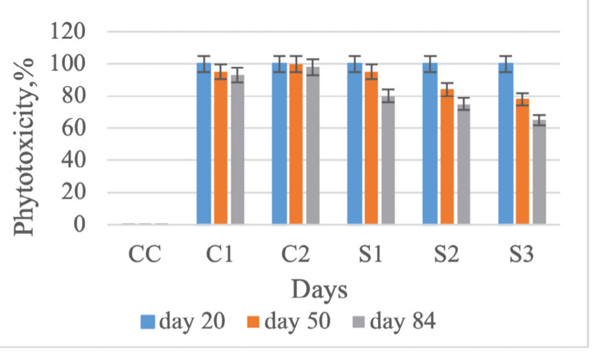

Figure 3. Influence of sorbents on the samples per day on Phytotoxicity percentage.

Testing for phytotoxicity was chosen because it is one of the most important indicators of the effectiveness of remediation. Graph 3 shows that in the pure CC control, phytotoxicity is zero, that is, the germination rate of crops is 100%. Nevertheless, in the samples with the addition of sorbents S1, S2, and S3, phytotoxicity gradually decreases, by day 84 in the S3 sample it reaches the lowest value compared to the rest. It is also clearly seen that in samples C1 and C2, the phytotoxicity practically does not change and remains 100%. Accordingly, according to this test, the work of sorbents in sorption bioremediation is visible. For a very important indicator – phytotoxicity, we have the following data. In CC control, phytotoxicity remains 0%, throughout the entire period. In sample C1 (without sorbent), phytotoxicity decreased from 100% (day 20), 95% (day 50) to 93% (day 84), and in sample C2 (without sorbent and biological product) from 100% (day 20), 100% (day 50) to 98% (day 84). These are very low indicators, and they demonstrate that the toxicity remains very high in samples without sorbents and biologics. But in samples with the addition of sorbents of various classes, the toxicity decreased more significantly. in sample S1, phytotoxicity decreased from 100% (day 20), 95% (day 50) to 80% (day 84), in sample S2, phytotoxicity decreased from 100% (day 20), 84% (day 50) to 75% (day 84) and the best result we have in sample S3: phytotoxicity decreased from 100% (day 20), 78% (day 50) to 65% (day 84).

Below are the photos that were taken during the experiment. Figure 4 shows photos of a sample with clean soil. The top photo shows the soil at the very beginning when the experiment was started. Here you can clearly see that the soil has its natural color, it is healthy and not contaminated with petroleum products. This soil sample was taken by us as a background sample. Below is a photo of the growing mustard seed Sinapis. The photo was taken 84 days after the start of the experiment. This photo shows the purity of the original soil, its fertility. Based on the results of germination in this sample, we evaluated such an indicator as to the phytotoxicity of the soil.

Figure 4. CC - clean control.

Figure 5 shows photos of samples of soil contaminated with oil and without the introduction of sorbent. According to these photos, the contaminated soil, in the absence of a sorbent, but even with the presence of a biological product, does not show a positive result after 84 days. Hence, it can be concluded that the use of an exclusively bacterial preparation does not have a positive effect on oil-contaminated soil and does not contribute to its bioremediation.

Figure 5. C1 - control without sorbent.

Figure 6. C2 - control without sorbent and biological product.

Figure 6 shows two photos of the soil, the first is initially contaminated with oil, it is not added to the sorbent and bioprerate. In this sample, we are interested in the process of self-purification of the soil from petroleum hydrocarbons. That is, we model a situation in which oil remediation and cleaning of the polluted territory are not carried out. In the lower photo, we can see that at the time of 84 days, after planting the phytotest, we have a small germination rate of plants. This means that the native microflora has become more active in the oil-contaminated soil, and as a result, a certain number of hydrocarbons has decomposed, and the soil has become less toxic. But the germination rate in this sample is quite low, so we can conclude that the natural microflora is not enough for self-purification and the introduction of sorbents is necessary. This is shown in the following photos.

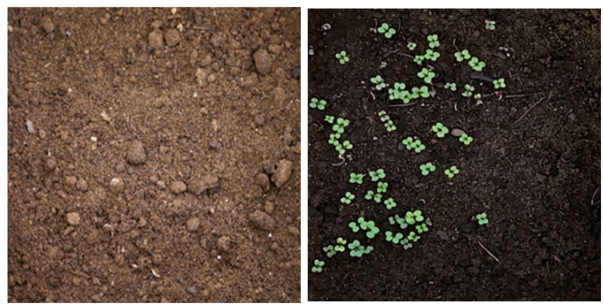

Figure 7. S1 - organic sorbent, peat.

Figure 7 shows photos of sample S1. In the upper photo, the sample, the soil, is polluted with noftu and with a peat sorbent applied. Below is a photo after sowing a plant crop. From the bottom photo, we can see that the presence of an organic sorbent significantly increases the germination rate of the plant.

Figure 8. S2 - mineral sorbent, vermiculite

Figure 8 shows photos with the sorbent vermiculite. Also, the bottom photo clearly shows a positive trend in the germination of the plant. This also demonstrates a decrease in soil phytotoxicity. At the same time, if we compare samples S1 and S2, we can see that the germination rate in sample S2 is higher, and, accordingly, the phytotoxicity of the soil is slightly lower.

Figure 9. S3 is a composite based on a carbon sorbent.

Figure 9 shows photos of the S3 sample. Here we demonstrate the work of the composite sorbent based on activated carbon developed by us. The lower photo clearly shows that the number of sprouted sprouts is higher than in samples S1 and S2. This effect is achieved due to the innovative composition of the sorbent developed by us. Activated carbon, which is present in the composition of the sorbent, actively sorbs petroleum hydrocarbons, due to reversible sorption, thereby actively reducing the toxicity of the soil. The upper photo was taken immediately after the sample was contaminated with oil, the lower one after 84 days. The photos clearly show the effect of sorbents. It is seen that samples S1 (fig. 7) and S2 (fig. 8) are approximately equally effective, but sample S3 (fig. 9) differs significantly in the appearance of sprouts, as well as in phytotoxicity data (Graph 3. Phytotoxicity values in samples). After 84 days, we have the following picture. In the CC (Clean Control) sample, the phytotoxicity is 0, while in the C1 and C2 samples, the phytotoxicity is quite high, significantly higher than in the samples with sorbents. Accordingly, it can be concluded that the use of sorbents reduces the toxicity in the samples.

Conclusions

- Sorption technologies are promising for isolation and use of natural microflora consisting of associations of microorganisms-destructors of directed action when creating biosorption complexes for cleaning and recultivation of soils contaminated with toxicants.
- The use of a composite sorbent based on carbon, mineral, and vegetable raw materials as a carrier for the immobilization of microorganisms allows us to create effective sorption and biological systems for the detoxification of hydrocarbons. The biocomplex must have its advantages in natural conditions such as ecological friendliness and biodegradability.
- The native microorganisms are well adapted to environmental conditions, being highly stable and synergistic, which allows for more efficient use of the target substance as the only food source.
- Based on the data of Fig. 1 it can be concluded that it is extremely important to monitor pH values in samples since the presence of petroleum products can acidify the soil, which negatively affects the work of oil-destroying microorganisms.
- The presence of sorbents, especially composite based on carbon (sample S3), increases the humidity in the samples. This has a positive effect on the rate of bioremediation, and consequently on the cleaning of the soil. At the same time, it is clearly seen that in the samples with the control of C1 and C2, the humidity is very low, from which it can be concluded that the presence of sorbents has a positive effect.
- From phytotoxicity testing, it can be concluded that the presence of sorbents of different classes has a positive effect on reducing toxicity in the samples, but the best result is shown by the S3 sample consisting of a composite material.

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