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ENVIRONMENTALLY SAFE RECLAMATION OF SOLID WASTE LANDFILLS

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Abstract. The main stages of the reclamation of solid waste landfills have been considered. The processes that limit ensuring the ecological safety of reclamation have been identified. In particular, three stages that may be hazardous for the environment have been highlighted: leachate treatment, preparation of soil or substrate for biological reclamation, and preparation of seeds and planting material. Leachate treatment by the aeration method in an aerated lagoon is recommended. The prospect of using such a process has been investigated in laboratory conditions. To ensure the prolonged release of nutrients in the reclamation layer, it is suggested to use encapsulated fertilizers. The effectiveness of the encapsulated fertilizer covered with a PET shell has been studied. For afforestation using high-quality planting material, the use of in vitro microcloning technology is proposed. The perspective of the method is shown in the example of microcloning of the white mulberry Morus alba L.

Keywords: reclamation, solid waste landfills, leachate, aerated lagoon, encapsulated fertilizers, substrate, microcloning.

1. Introduction

Used solid waste landfills are ecologically dangerous objects that impact soil, subsoil, atmosphere and hydrosphere. The problems of reclamation of these objects, which do not meet the norms of environmental safety, are essential for many countries whose anthropogenic activity for many years has not been environmentally balanced and safe. Ukraine belongs to such countries. Today, the task of Ukraine, which has declared its political direction – joining the European Union and is taking confident steps in this direction, is not only the harmonization of state legislation (including environmental protection) with the legislation of the European Union but also the harmonization of technologies, practices, mechanisms and strategies in the field of safe anthropogenic activity. From the point of view of environmental safety, this corresponds to the Law of Ukraine "On Waste Management" (Zakon Ukrainy, 2022) adopted by the Verkhovna Rada of Ukraine, which in its main provisions is harmonized with the EU Directive "On Waste" 2008/98/EC (Eur-Lex, 2008). By this Law, National (and based on it, regional) waste management plans are developed. The National Waste Prevention Program is a component of the National Waste Management Plan. According to Chapter XI of the

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Law of Ukraine "On Waste Management" "from January 1, 2030, is forbidden to operate waste disposal sites (landfills) that are not equipped with biogas and leachate extraction and disposal systems, systems for monitoring emissions into the atmosphere and monitoring soil and underground pollution waters... The owner (balance keeper) of the landfill or the business entity managing the landfill should ensure the reclamation of the landfill after the termination of its operation, as well as its care after its reclamation for 30 years". These studies are directed to the development of scientific and practical foundations of environmentally safe processes in the technologies of reclamation of waste disposal sites.

According to DBN V.2.4-2-2005 "Solid Waste Landfills. Basic Design Provisions" (Polihony tverdykh pobutovykh vidkhodiv, 2005), land reclamation after the closure of a solid waste landfill is carried out in two stages: technical and biological. The territory of reclaimed lands of a solid waste landfill, according to the DBN, is transferred to the relevant department for the following targeted use in agriculture, forestry or other areas. However, to implement both stages of reclamation, a large number of stages and processes are used, which are closely interrelated. Among these processes, there is a significant number of technical and mining operations similar to those implemented in the arrangement or reclamation of spent mining chemical or construction quarries (mining operations, laying out the landscape, creating a protective screen and fertile layer, etc.), which is determined by the conditions of ensuring civil security (Fig. 1).

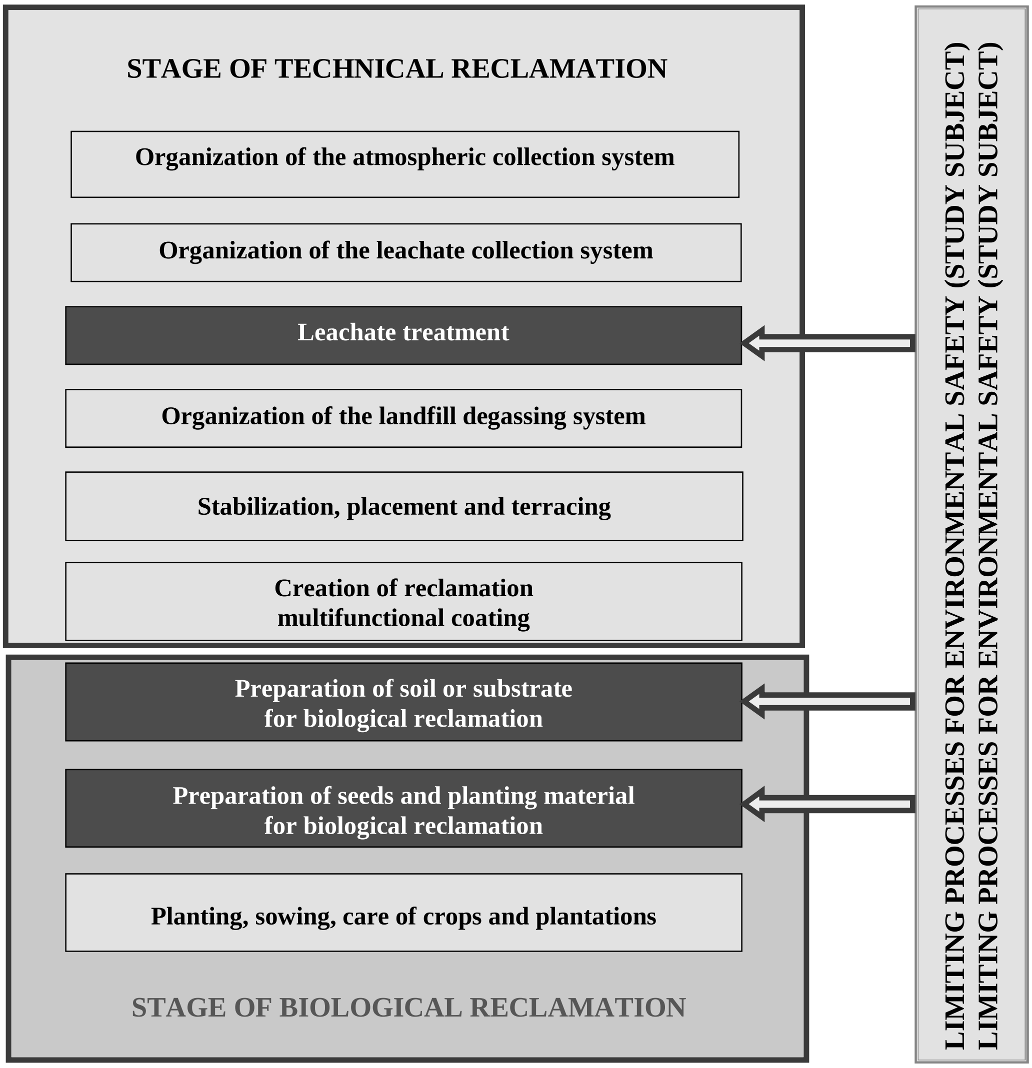


Fig. 1. Structural scheme of the reclamation processes of solid waste landfills

The subject of research is the processes that determine the ecological safety of reclamation, which must be applied both at the stage of technical reclamation and at the stage of biological reclamation. At the stage of technical reclamation, the treatment of leachates determines environmental safety and prevents the pollution of the hydrosphere. To solve the problem of elimination of environmental hazards caused by leachates of landfills at the stage of their closure, it is necessary to highlight two stages: 1 – treatment of the accumulated leachates to enable the implementation of the reclamation project of the solid waste landfill in the future; 2 – treatment of leachates, which after the reclamation of the solid household waste landfill will be formed in the body of the landfill for decades as a result of biological processes of decomposition of the organic component of garbage. These stages fundamentally differ in terms of the volume consumption of leachates that will be sent for treatment, physicochemical characteristics and duration of implementation of each of the stages. It is ineffective to provide a single technology for the implementation of both these stages. The reasons for this are technological (impossibility of ensuring full load and efficient operation of equipment) and financial (significant cost overruns) aspects.

The following technologies recommended in Ukraine (Yakist vody, 2012) became the most widespread: reverse osmosis technology (Dushkyn et al., 2011); technology of electroplasma treatment of leachate (Mashal et al., 2012; Gomez et al., 2009); evaporation and drying technology (Yakist vody, 2012); biological treatment technology in anaerobic (Dzhamalova, 2015) and aerobic (Malovanyy et al., 2018) environments. All these technologies have both advantages and disadvantages, and the expediency of their use in each specific case is determined based on technical and economic analysis of a specific situation. But in all cases, it is advisable to use a two-stage treatment strategy. At the first stage of preliminary treatment, it is advisable to use a financially inexpensive technology that would allow leachate treatment from at least half of the impurities and thus reduce the load on the second, final stage, at the output of which the leachate should meet the standardized indicators in terms of the content of impurities. The first stage could be the preliminary aerobic biological treatment of the leachate in the conditions of an aerated lagoon.

As for the preparation of the soil or substrate, which creates a fertile layer on the surface of the film-insulated body of the landfill (and which is an important aspect of achieving the ecological safety of the reclaimed landfill), it is important to provide it with a balanced set of nutrients that are necessary for the successful implementation of the biological stage of reclamation. A promising method of releasing nutrients from fertilizer particles into the soil solution is to cover the mineral fertilizer particles with capsules using polymers (encapsulated mineral fertilizers). It is promising to create polymer capsules that cover fertilizer granules. Encapsulation is enclosing small amounts of materials or substances in a capsule (shell) to obtain closed capsules with specified properties. As a result of encapsulation, the isolation of a particle of the substance (base substance) from the environment in which the capsule is placed, as well as the isolation of individual particles of the base substance, is achieved. The constituent particles of the capsule are called a core and a shell. The method of isolating the core particles both from the external environment and other similar particles is to create a diffusion resistance of the shell, which either makes the interaction of the core with the external environment impossible or significantly complicates it (Gumnitsky et al., 2006). This slowdown in the release of nutrients from the capsule is determined mainly by the thickness and composition of the polymer capsule. When the capsules are impermeable but capable of biodegradation in the soil environment, the contents of the granule are released into the soil environment as this biodegradation occurs. For this option, the release is regulated not by the intensity of dissolution of the mineral fertilizer granule but by the beginning of the biodegradation of the capsule (Gospodarenko, 2002; Nahurskyi, 2012).

Another significant aspect of reclamation, which ensures its success and ecological safety, is the problem of preparing high-quality planting material for the afforestation of territories at the stage of biological reclamation and reproduction of woody plants in controlled artificial conditions *in vitro*. The preparation of planting material for the propagation of woody plants under controlled artificial conditions *in vitro* will make it possible to use only high-quality, selectively valuable planting material in biological reclamation technologies, which will significantly enrich the gene pool and increase the stability and productivity of stands. This method of obtaining planting material eliminates the dependence on the periodicity of seed years and yield, as well as seed quality and environmental parameters. In microcloning, several successive stages are distinguished, which were studied by some researchers. For example, there are studies on the decontamination and initiation in the reproduction of *Morus alba L. in vitro* by scientists (Agarwal et al., 2004; Ahmad et al., 2007) and the multiplication of explants of the same culture (Balakrishnan et al., 2009; Barbosa et al., 2005). Studies have been devoted to the problems of microcuttings rooting and adaptation of *ex vitro* regenerants (Anis et al., 2003; Benedetta et al., 2007).

The purpose of the research was to establish the perspective of using the processes and methods listed above to ensure environmentally acceptable reclamation of solid waste landfills and to prevent the possible negative impact of reclamation processes on the environment.

2. Experimental part

Ammonium nitrogen is one of the most dangerous contaminants of landfill leachates. Therefore, the change in the concentration of ammonium nitrogen during the treatment process served as a criterion for assessing the prospects of using the aerated lagoon method for leachate treatment. The concentration of ammonium ions was determined using a known method (Yakist vody, 2012), according to which the method of calibration graphs was used to determine the concentration of ammonium nitrogen. Before constructing the calibration graph, a series of 10 solutions was prepared, in which light absorption was measured (wavelength λ=425 nm). Calibration graphs were built for each new preparation of the necessary reagents (ferret salt and Nessler's reagent). Research on the leachates treatment was carried out under conditions of constant aeration of the investigated leachates, which was implemented in a laboratory 5-litre thermostated flask (model of an aerated lagoon) with the help of a laboratory compressor. Samples were periodically taken from the flask and analyzed for the content of ammonium nitrogen. The leachates from the Lviv (Hrybovychi) solid waste landfill and the Chervonohrad (Lviv region) solid waste landfill were used for the research. Due to the long period of operation of the Lviv landfill (since 1958), leachates from this landfill are a typical representative of the so-called “old” leachates. At the same time, the majority of solid waste landfills (including Chervonohrad) are operated for an average of 10–30 years, and leachates from such landfills are usually classified as “young” leachates, which are characterized by different ratios of the content of the main pollution indicators

In the studies on the prospects of using encapsulated fertilizers in reclamation technologies, it is proposed to use modified polyethyl terephthalate (PET) as a material for capsules. Huge volumes of use of PET products, mainly in the form of containers, lead to massive pollution of the environment with this plastic. The existing methods of disposal of PET waste are not able to ensure their complete disposal. It is appropriate to use such waste as secondary material resources since the properties of plastic practically remain unchanged during the use of products. For PET, there is a system of separate collection (used PET bottles) and disposal (secondary raw materials for the production of fibre, PET bottles, sheets for thermoforming, strapping tape, etc.). We investigated the possibility of modifying PET by implementing an alcoholysis reaction using diethylene glycol as a reagent. As a result, the solubility of modified PET in ethyl acetate is achieved, which is sufficient for the technological process of capsule formation in a fluidized bed apparatus. PET waste in the form of flakes, which underwent initial processing at a specialized enterprise, and diethylene glycol (DEG) in a PET: DEG molar ratio of 1:0.5, which were loaded into a hermetic reactor, were used for research. The contents of the reactor were heated to a temperature of 493 K. Two hours after reaching the required temperature, the vacuum pump was turned on, and ethylene glycol was distilled from the reactor at a residual pressure of 20 kPa. The total duration of the process was 3.5 hours. As a result of the interaction, ethylene glycol is displaced by diethylene glycol to obtain a product soluble in ethyl acetate. This solution was later used for encapsulation on particles of mineral fertilizers (azofoska and ammonium nitrate) in a fluidized bed apparatus.

Determination of the concentration of nutrients at any time during the dissolution of encapsulated mineral fertilizers was carried out by the conductometric method, which is based on the measurement of the electrical conductivity of the solutions. The electrical conductivity of the solution was measured in a conductometric installation, which consists of a measuring electrode, the non-working surface of which was insulated with a dielectric. The electrode was fixed in the lid of the measuring flask with distilled water. Levelling of the concentration of the solution at the time of measurement was carried out using a magnetic stirrer. To determine the concentration of the target component in the solution during the extraction process, the device was calibrated with standard solutions of the research objects at a temperature of 293K. Based on the obtained data, a calibration curve was constructed. After linear optimization of the experimental data of the electrical conductivity of the solution, we obtained the equation of the dependence of the component concentration in the C1 solution on its electrical conductivity. According to the obtained equation, the current concentration of mineral fertilizers in the solution was calculated when studying the release kinetics.

White mulberry *(Morus Alba Linn.)* was used to test the use of micro-cloning to obtain promising planting material for biological reclamation of solid waste landfills. White mulberry is a valuable exotic tree, naturalized in Ukraine due to its high fruit, medicinal and decorative properties. The collection fund of mulberry, represented in Ukraine, includes more than a hundred zoned, high-yielding and biologically resistant selection, and decorative forms. In the study of the peculiarities of *in vitro* cultivation of white mulberry (*Morus Alba Linn*.), 3-year-old seedlings and 50-year-old specimens of white mulberry acclimatized in the conditions of the Botanical Garden of the National Forestry University of Ukraine were donors of explants for *in vitro* reproduction. Shoots were selected three times during the growing season: before the beginning of the growing season (March), at the beginning of the growing season (April) and during the most active growth of shoots (June). Apical and lateral buds were used as explants to obtain growth cones. Explants were marked with the first letters of the species' name, indicating the age of the donor and the time of selection. For example, Ma3/4, where 3 is the age of the donor (years), and 4 is the month of selection (April). Chemotherapy of explants consisted in performing the following consecutive operations: washing with running water with the addition of household soap for 2–3 hours; alternating treatment with antiseptic preparations: ethanol (96 %) – 3 seconds, sodium hypochlorite (1.5 % in terms of active chlorine) – 10 minutes, silver nitrate (0.1 %) – 15 minutes. After treatment with each agent, the explants were washed three times with sterile distilled water for 3–5 minutes. After surface decontamination, the explants were disassembled in a laminar box to get growth cones of 2–3 mm in size and aseptically transferred to solid nutrient medium MS without phytohormones for 12 days. As part of the experiment, undissected buds, selected in April (marked by n), were also passaged on the medium. For each separate experiment, 60 explants were used. *In vitro* thermotherapy consisted in cultivating explants at a temperature of 38±1 °C, treatment exposure - the first 4 days (1/3 of the cultivation time) and a 14-hour photoperiod. Given the positive results of decontamination after chemotherapy and thermotherapy, chemotherapy was not used.

3. Results and Discussion

3.1. Study of the prospects of leachate treatment by the aerated lagoon method

To compare the effectiveness of the biochemical treatment of "old" and "young" leachates, we built dependencies using data obtained in the biochemical leachate treatment of the Chervonohrad landfill ("young" leachate) and data of the biochemical treatment of the leachate of the Hrybovychii (Lviv) landfill ("old" leachate). The comparison was carried out by means of a comparative analysis of the kinetics of biochemical aerobic purification of both types of leachates from ammonium nitrogen. The results of the research are presented in Fig. 2.

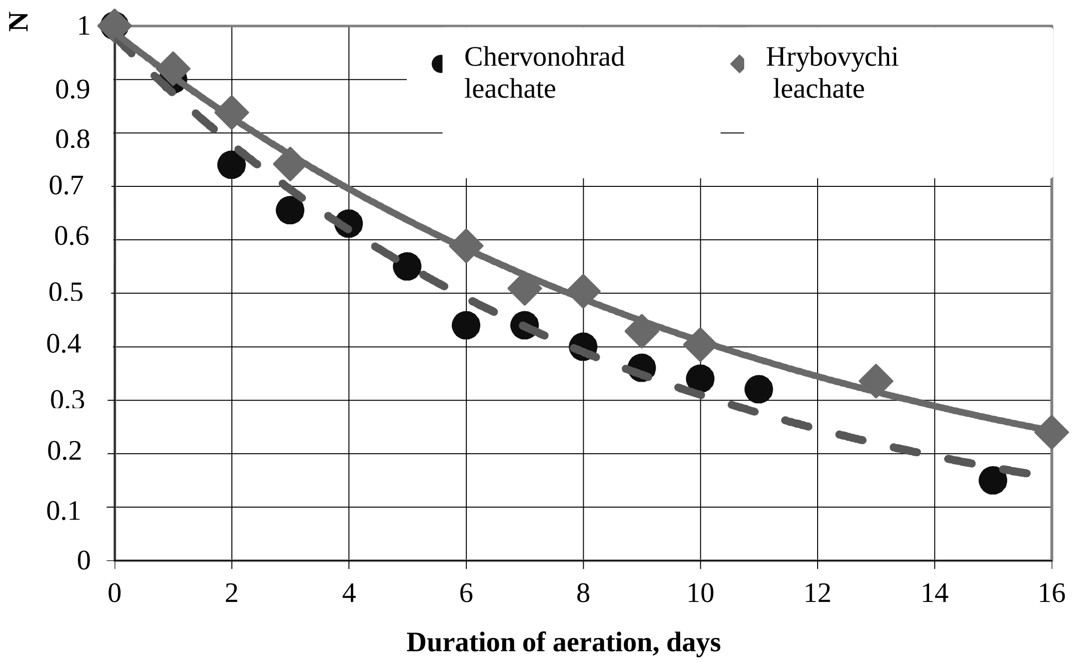


Fig. 2. Comparative analysis of the kinetics of changes in the relative concentration of ammonium nitrogen (N) in Hrybovychi and Chervonohrad leachates

To ensure the correctness of the comparison, the indicator "relative concentration of ammonium nitrogen in the leachate (N)" was used, which was determined by the formula:

$$N = \frac{C(NH_4 - N)_{in}}{C(NH_4 - N)_{fin}}, \quad (1)$$

where $C(NH_4\text{-}N)_{in}$ and $C(NH_4\text{-}N)_{fin}$ are, respectively, the initial (fixed at the beginning of the treatment process) and final (for a fixed period of treatment) concentration of ammonium nitrogen in the leachate which is being treated.

Fig. 2 shows that for both types of leachate, the treatment of ammonium nitrogen is implemented by approximately the same mechanisms, and both groups of results can be described by exponential dependencies. Approximation equation for changing the relative concentration of ammonium nitrogen in the Hrybovitsky leachate ($R^2 = 0.9947$):

$$N = 0.9874e\text{-}0.0877x, \quad (2)$$

where N is the relative concentration of ammonium nitrogen; x is the duration of aeration, days.

The approximation equation for the change in the relative concentration of ammonium nitrogen in the leachate of the Chervonohrad landfill (R2 = 0.9759):

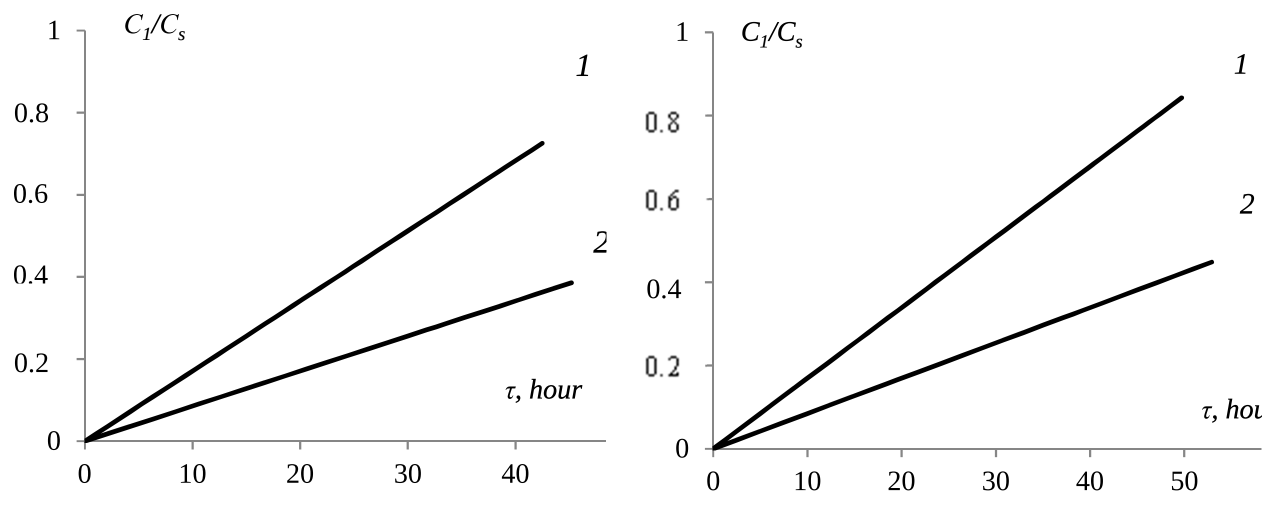
$$N = 0.9807e\text{-}0.1152x. \quad (3)$$

As can be seen from the data shown in Fig. 1, the oxidation of ammonium ions from "young" leachate is more intensive, moreover, the amount of oxidized ammonium ions in the case of treatment of "young" leachates (1427 mg/l → 212 mg/l) is much greater than the same ratio in the treatment of "old" leachate (899 mg/l → 219 mg/l).

3.2. Study of the prospects of the use of encapsulated mineral fertilizers for biological reclamation

After obtaining encapsulated mineral fertilizers by the method described above, studies were conducted on the prospects of their use in reclamation technologies. In particular, an important indicator of the quality of the applied coating on a particle of basic fertilizer is the uniformity of the shell thickness. This makes it possible to predict the intensity of the release of components into the soil environment and, accordingly, to create encapsulated fertilizers with the required duration of action.

According to the obtained technological parameters, the encapsulation of granular fertilizers (ammonium nitrate and azofoska) was carried out in a cylindrical apparatus of periodic action. The amount of coating was 10 % and 20 % of the mass of fertilizers, which corresponds to the following average thicknesses of the film on the surface of the particles (10^{-5} m): ammonium nitrate – 5.74 and 11.48; azofoska – 5.23 and 10.46. The solubility of the obtained fertilizers was checked experimentally by the conductometric method. Encapsulation quality control was monitored by the nature of the release curve. The results of the research are shown graphically in Fig. 3.



a) ammonium nitrate b) azofoska

Fig. 3. Dissolution kinetics of encapsulated fertilizers covered with a shell based on modified PETF of different thicknesses (% by weight): 1 – 10, 2 – 20

The obtained results (Fig. 3) indicate that the kinetic curves of dissolution have a predictable character, and the process proceeds smoothly without sharp declines or rises. This confirms a uniform, high-quality coating, which makes it possible to obtain long-acting mineral fertilizers with the necessary release time. It is precious for the implementation of biological reclamation of solid waste landfills.

3.3. Study of the prospects of introducing white mulberry (*Morus Alba Linn.)* into the culture *in vitro*

As can be seen from Fig. 4, the applied technology made it possible to obtain a high proportion of white mulberry (*Morus Alba Linn.*) decontamination, M.

The success of obtaining a healthy initial explant depends on its genotype, age, place and time of selection and the correctly selected decontamination method. Fig. 3 shows that the best result was achieved during processing of explants Ma3/4 (98.3 %) and Ma50/4 (91.7 %), selected at the beginning of the growing season (Fig. 5).

Given the high percentage of necrotic explants (Ma3/4 n and Ma50/4 n – 76.7 and 95.0 %, respectively), decontamination and cultivation of undissected buds proved ineffective. Explants selected during the most active growth of shoots had the highest percentage of contamination (Ma3/6 and Ma50/6-25.0 and 33.3 %, respectively).

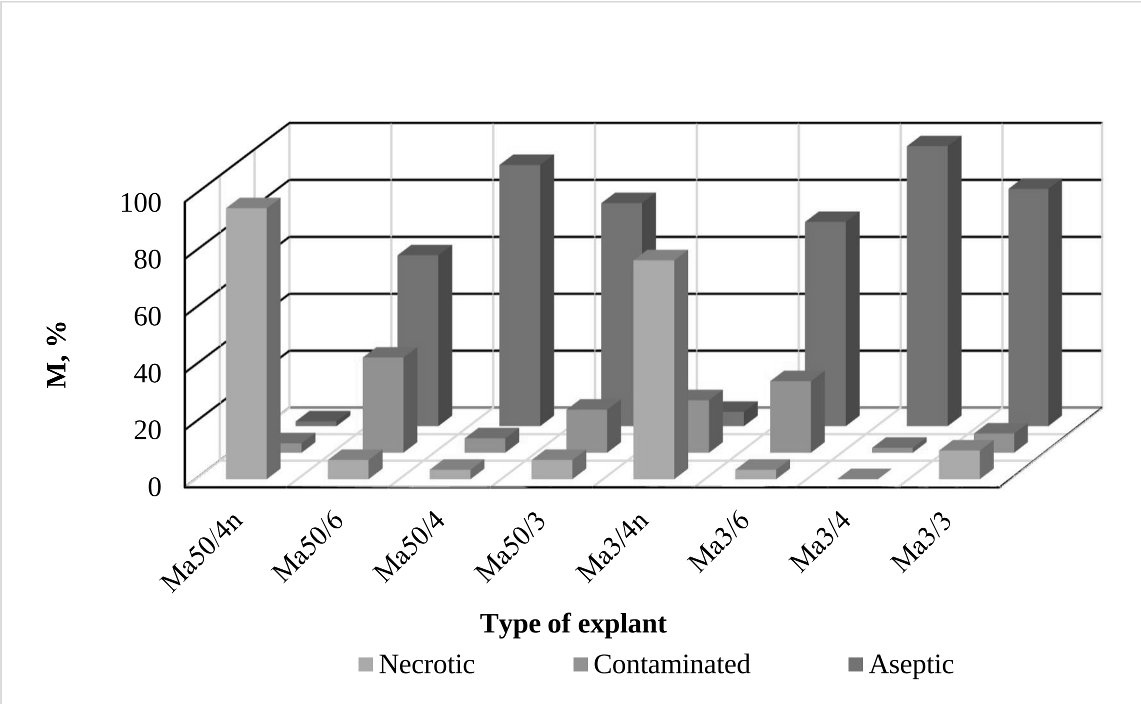


Fig. 4. Diagram of decontamination of different types of white mulberry explants

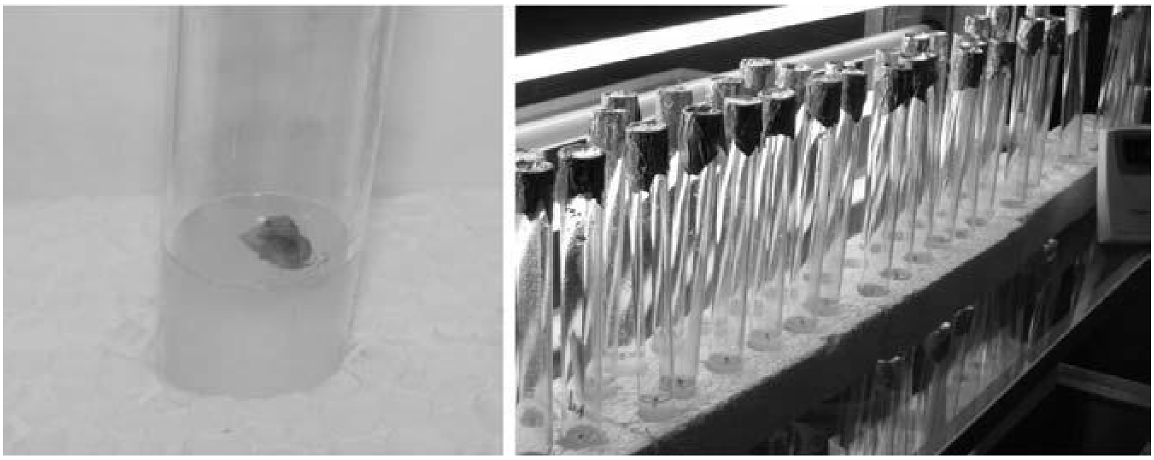


Fig. 5. Aseptic explants of white mulberry

4. Conclusions

According to the results of laboratory studies of the leachates of the Lviv (Hrybovychi) and Chervonohrad landfill sites (Lviv region), the promising application of the aerated lagoon method for treating all types of leachates is shown. This makes it possible to recommend this type of treatment as the first stage (pre-cleaning) in treating leachates of all types of solid waste landfills. It is advisable to carry out the final treatment using reverse osmosis or according to existing technology at sewage treatment plants or artificially created wetlands.

Test studies of encapsulated fertilizers were carried out according to the EN 13266:20 methodology. The obtained results indicate that the kinetic curves of dissolution have a predictable character, and the process is implemented according to monotonic dependencies. This confirms the uniformity and quality of the coating, which makes it possible to obtain and use encapsulated mineral fertilizers of prolonged action with the necessary release time for biological remediation. This is important for biological reclamation because there is no need for frequent agrochemical operations, and there is no danger of damage to the reclamation layer.

During the introduction of white mulberry acclimatized in Ukraine into in vitro culture as explants, the best result was achieved due to the selection of buds of juvenile plants at the beginning of the growing season. In the case of decontamination, the highest share of aseptic explants (98.3 %) was obtained after chemotherapy and thermotherapy. This allows us to assert the prospects of using microcloning to prepare high-quality planting material for the reclamation of spent solid waste landfills through afforestation.

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