ENVIRONMENTAL PROBLEMS

Vol. 6, No. 4, 2021

PROSPECTS OF CLEANING FILTRATES BY AEROBIC-REAGENT METHOD ON THE EXAMPLE OF LVIV REGION

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https://doi.org/10.23939/ep2021.04.264

Received:27.10.2021

Abstract. In 2021, the situation with the collecting, processing and disposal of waste in general, solid waste in particular, remains difficult in Ukraine and needs to be solved immediately. Soil contamination, as well as contamination of surface and ground waters by filtration effluents (or filtrates), is one of the most dangerous consequences of non-regulatory disposal of solid household waste.

Keywords: solid household waste, filtrates, aerobic biochemical treatment, reagent treatment.

1. Introduction

In 2021, the situation with the collecting, processing and disposal of waste in general, including municipal solid waste (MSW), remains problematic in Ukraine and needs to be solved urgently. In 2019, 441.5 million tons of waste were generated in Ukraine, which is 20.2% more than in 2018, with 98.7 % (435.6 million tons) of waste generated as a result of economic activity, including 390.6 million tons (88.5 % of the total amount) - in the extractive industry and quarrying, as well as 1.3 % (5.9 million tons) - in households.

According to official data, the total amount of solid waste collected in Ukraine in 2019 was 52.9 million m^3 or about 10.4 million tons (KND 211.1.4.030-95, 1995), that corresponds to a specific generation of solid waste of 140 kg/(man × year). At the same time, a very small share of solid waste processing and utilization remains typical in Ukraine: in 2019, 49.7 million m^3 or 9.8 million tons of solid waste were disposed at landfills and dumps, which is about 94.2% of the amount of collected solid waste.

One of the most dangerous consequences of non-regulatory disposal of solid waste is the contamination of soil, as well as surface and ground water by filtration effluents (or filtrates). Based on the range of the above mentioned solid waste storage and disposal areas, with an average annual precipitation layer height of 500 mm/year and runoff values 0.05−0.1, from 2.2 to 5.0 million m^3 of filtrates is formed annually at landfills and dumps of solid waste in Ukraine. Depending on the degree of dilution by atmospheric water, the concentration of the main pollutants in the filtrates is 5–50 times higher than the limit values. The lack of waterproofing layers, drainage systems, collection and treatment of filtrates at many landfills and dumps let highly toxic filtrates get directly into soil, surface and ground water, causing great, often unpredictable damage difficult to quantify. It also directly affects the health of residents of adjacent areas.

The largest leachate volumes (about 100 thousand m^3 and more) were collected in storage ponds at the largest landfills in Ukraine: at the Kyiv landfill №5 and the Lviv landfill in V. Hrybovychi, Lviv region in particular.

Typical landfills and dumps of medium capacity, which are geographically dispersed, are a dangerous source of local pollution in hundreds of settlements throughout Ukraine. They pose serious a threat to the environment and human health. According to the official statistics (KND 211.1.4.030-95, 1995), the average area of a landfill in Ukraine is about 1.5 hectares, which corresponds to the estimated average annual amount of newly formed filtrate in the range of 0.38-1.1 thousand m^3

For citation: Malovanyy M., Zhuk V., Tymchuk I., Vronska N., Zavoyko B., Senkovych O., 2021. Prospects of cleaning filtrates by aerobic-reagent method on the example of Lviv region. Journal Environmental Problems. Vol. 6, No. 4. p. 264–269. DOI: https://doi.org/10.23939/ep2021.04.264

of filtrates at the height of the annual rainfall level of 500−700 mm/year and runoff coefficient values 0.05−0.1.

The vast majority of landfills and dumps in Ukraine do not have filtrate treatment systems, and many such facilities do not even have waterproofing and filtrate collection systems, which makes the problem of filtrate collection and treatment particularly relevant.

The aim of the research was to develop technologies for pre-treatment of landfills and solid waste landfills (MSW), which will allow sending pre-treated filtrates for additional treatment to municipal sewage treatment plants.

2. Methods and techniques

Aerobic methods of biological filtrate treatment have some indisputable advantages over anaerobic ones: they are flexible in use, quickly enter the stationary mode of operation, quickly adapt to the variable composition and consumption of infiltrates. Aerobic reactors are much simpler in design and much cheaper than anaerobic ones. They are also easier to automate and operate.

The technology of pre-biochemical leachates treatment in aerated lagoons is especially promising for implementation at landfills and waste disposal sites in Ukraine (Calli et al., 2006; Maehlum, 1995; Mehmood et al.,2009; Robinson, Grantham, 1988; Sawaittayothin, Polprasert, 2007). Microbial communities, which are present in facilities for biological wastewater treatment, are well adapted to the destruction of complex organic compounds in the streams of various liquid wastes, including landfill and waste disposal sites filtrate. In the system of landfill filtrate treatment, the corresponding aerobic biocenosis occurs. It can efficiently oxidize complex organic compounds of highly concentrated filtrates.

Reagent methods of filtrate treatment, coagulation and flocculation, in particular, are widely used for preliminary or complete purification of landfill filtrates. These methods allow removing biologically non-oxidizing humic and fulvic acids, as well as other specific "heavy" contaminants, including heavy metal ions, organochlorine compounds and the like from the filtrates.

Fenton method (and its modifications) is one of the most effective methods of landfill filtrate reagent treatment. The classical Fenton reaction is a chemical reaction of hydrogen peroxide decomposition in the presence of iron salts, accompanied by the formation of active radicals that trigger radical-chain transformations, including biochemical ones, which are capable of destroying biological compounds:

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + OH^* \quad (1)$$

Systematic investigations have been conducted during the last two decades on the possibilities of Fenton process and its varieties (photo-Fenton process, electro-Fenton process) application in the schemes of preliminary or complete treatment of landfill filtrates (Badawy et al., 2013; Bae et al., 1997; Deng, Englehardt, 2006; Wiszniowski et al., 2006).

Chervonohrad landfill filtrate was used for laboratory research as a typical "young" filtrate of Ukrainian landfills.

The filtrate was selected in accordance with the requirements of relevant industry standards (DSTU ISO 5667-2-2003, 2003; DSTU ISO 5667-10-2005, 2005; KND 211.1.0.009-94, 1995). The procedure for filtrate selecting from the collector is shown in Fig. 1. Sample transportation and its storage in the laboratory of the Department of Ecology and Sustainable Nature Management of Lviv Polytechnic National University was carried out in accordance with the requirements (DSTU ISO 5667-3-2001, 2001).

Fig. 1. Filtrate selection for laboratory tests from the filtrate collector at the Chervonohrad landfill

Aerobic biochemical filtrate treatment. In the laboratory, the efficiency of pre-purification of the filtrate was investigated according to the main indicators of contamination: reaction of the medium (pH), biochemical oxygen demand (BOD), chemical oxygen (COD), total nitrogen content (the sum of organic and ammonium nitrogen), the content of suspended solids and surfactants (SS). In addition to the main technological parameters of the filtrate quality at different stages of its treatment, there were determined additional ones, namely, biochemical oxygen consumption for the first 5 days (BOC) and ammonium nitrogen concentration (NH4 +).

Studies of aerobic biochemical filtrate purification were performed in a laboratory on a specialized experimental plant (Fig. 2), which allows modelling the conditions of aerobic biochemical purification of the landfills and waste disposal sites filtrate in industrial conditions.

Laboratory studies of aerobic filtrate treatment were performed using the following method.

In a glass laboratory flask with a capacity of 5 dm^3, 4 dm^3 of the typical raw filtrate was poured, which was typical for small and medium landfills of Ukraine with a service life from 10 to 30 years. Chervonohrad landfill filtrate was taken from the collector as a typical one.

Air with a volume flow rate of 0.05 dm^3/s was supplied to the lower part of the flask from the laboratory compressor through the laboratory aerator. Air flow for aeration was measured with a laboratory rheometer and controlled by the position of the control valve.

Fig. 2. Scheme of an experimental plant for studying the efficiency of biological aerobic treatment of the landfill filtrate in aeration conditions

Initial, intermediate and final values, which were selected after certain periods, the main and auxiliary indicators of filtrate contamination were determined by the following methods:

– The value of COD was determined according to the national standard of Ukraine DSTU (National Standard of Ukraine) ISO 6060: 2003 (DSTU ISO 6060: 2003, 2003).

– The concentration of ammonium nitrogen was determined by the photometric method of ammonium ions with Nesler's reagent in wastewater KND 211.1.4.030-95 (KND 211.1.4.030-95, 1995). A photoelectric colorimeter brand KFK-2-IHL4.2 was used

– Method of determining the biochemical oxygen demand after n days (BOD) in natural and wastewater (KND 211.1.4.024-95), which accordingly determined the value of BOC_5 KND 211.1.4.024-95, 1995).

– The hydrogen pH of the test filtrates was determined using a pH meter Checker 1 HI98103 according to the relevant instructions.

– pH was determined potentiometrically using a pH meter Checker 1 HI98103.

Aeration was carried out in a continuous mode. Studies of filtrates treatment were performed at a constant study temperature of 20 ± 1^o C. The aim of the research was to establish the maximum effect of filtrate treatment that can be achieved in the process of aerobic biochemical oxidation in the laboratory conditions.

Reagent treatment of filtrates. The method of filtrates reagent treatment using a new modified method of Fenton, the basis of which was provided in (Malovanyy, Gorbach et al., 2018; Malovanyy et al., 2018), presupposes sequential introduction into a container of pre-biochemically purified filtrate of aqueous solutions of four reagents into a container. These solutions are: polyacrylamide or its more effective derivatives as a flocculant, an aqueous solution of aluminum sulfate as a coagulant and, further, simultaneously – solutions of iron (II) sulfate and hydrogen peroxide (Fenton's reagent). Thus, the whole complex of the four above reagents is one of the variants of the modified Fenton method. The idea of this modified method is the preliminary introduction into the filtrate of flocculant (PAA) and aluminium coagulant with the following addition of Fenton's reagent.

The volume of pre-biochemically purified filtrate in all experiments was the same and equal to W_f = 200 cm^3. Laboratory modelling of reagent treatment was performed in laboratory glass cylinders with a volume of 500 cm^3.

3. Results and Discussion

Aerobic biochemical treatment of the filtrate. Initial parameters of Chervonohrad landfill filtrate are: pH - 9.8; concentration of ammonium nitrogen - 1427 mg/dm^3; nitrogen concentration (sum of organic and ammonium nitrogen) - 2253 mg/dm^3; chemical oxygen demand - 5768 mg O_2/dm^3; BOC_5 - 128.5 mg O_2/dm^3; BOD_{total} - 507.5 mg O_2/dm^3; content of suspended solids and surfactants - 228 mg/dm^3.

In Fig. 4-9, the study results on the experimental plant are shown.

According to Fig. 3–9, the concentration of ammonium nitrogen changes from 1427 mg/dm^3 to 212.6 mg/dm^3 (6.7 times), the nitrogen concentration decreases from 2253 mg/dm^3 to 358, 2 mg/dm^3 (6.3 times) in the process of biochemical aerobic filtrate treatment. The value of COD decreased by only 30%, the filtrate needs further purification.

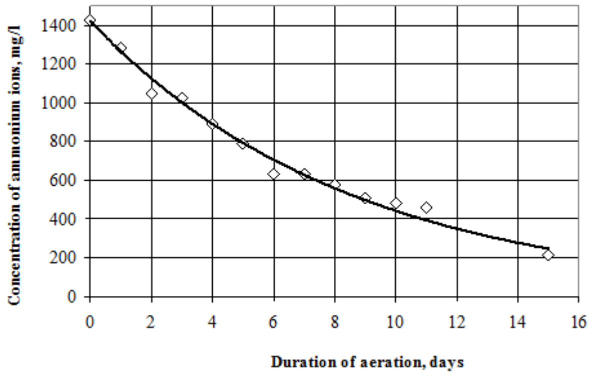

Fig. 4. Change in the concentration of ammonium nitrogen during aeration

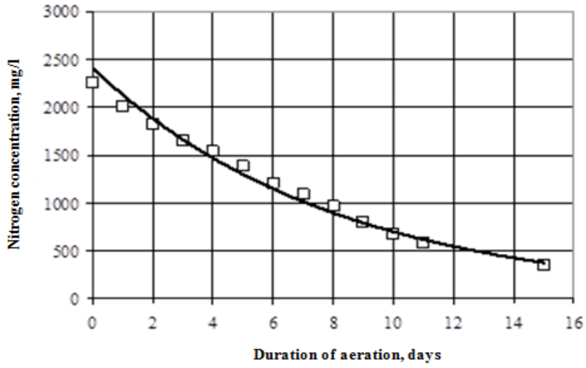

Fig. 5. Change in nitrogen concentration (the sum of organic and ammonium nitrogen) in the process of biochemical aerobic filtrate treatment

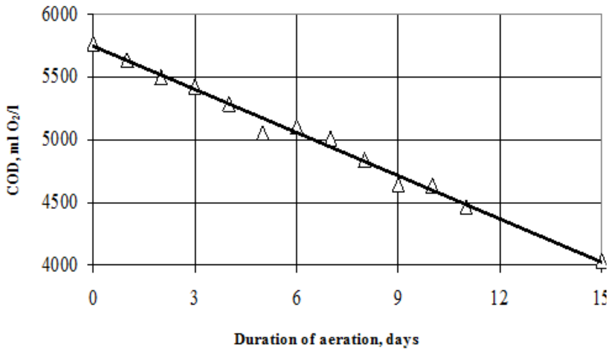

Fig. 6. Change of COD in the process of biochemical aerobic filtrate treatment

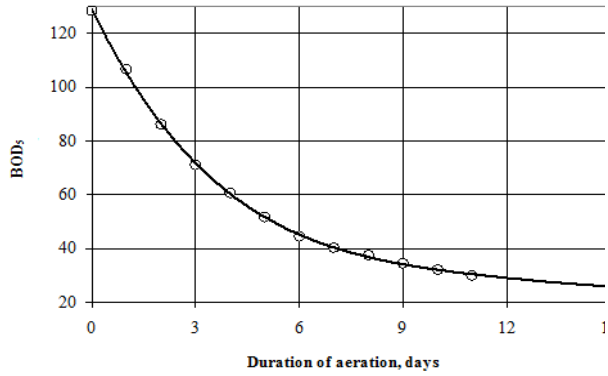

Fig. 7. Change of BOD_5 in the process of biochemical aerobic filtrate treatment

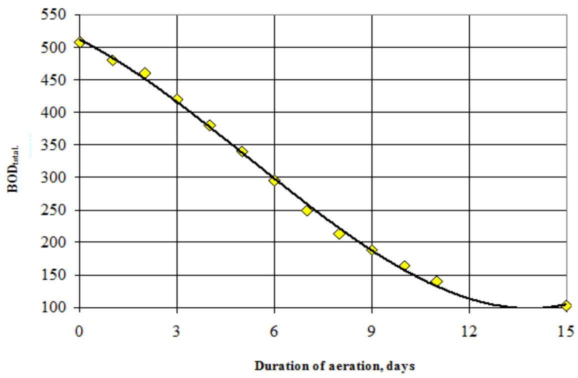

Fig. 8. Change of $BOD_{total.}$ in the process of biochemical aerobic filtrate treatment

Fig. 9. Change of pH during biochemical aerobic filtrate treatment

In the first period of aeration, there is a sharp decrease in the level of BOD_5, which is probably associated with the oxidation of easily oxidizable biological compounds. Then, the oxidation of biological contaminants slows down. In general, during the experiment the level of BOD_5 contamination decreased 5 times.

The dependence of filtrate pH change during aeration is random and is apparently determined by the nature of biochemical processes that take place during the filtrate oxidation by aerobic microbiocynosis which is inactivated in the purification reactor. Throughout the process, the pH value did not exceed the range of 9.6–10.2.

Reagent filtrates treatment. Optimal doses of working solutions of reagents were determined in the laboratory using pre-biochemically purified filtrate. The criterion for the optimal composition of working solutions were the initial values of filtrate contamination basic indicators after reagent treatment which, according to the requirements for the technical task to perform HTP, should not exceed the corresponding GN to be discharged into a centralized sewerage system in Ukraine (Order of the Ministry…, 2017).

The general tendency to decrease pH value, ammonium and total nitrogen concentrations, BOD and HSC with increasing doses of reagents was obtained.

The compositions of reagent solutions, which showed the greatest efficiency as a result of experimental studies, are given in Table 1.

Table 1

Reagent compositions in laboratory studies of filtrates reagent purification (filtrate volume W_f = 200 cm^3)

	Volume of aqueous solvents, cm^3			
	PAA (0,1% wt.)	Aluminum sulfate (10% wt.)	Iron sulfate (II) (10% wt.)	Hydrogen peroxide (10% wt.)
1	20	20	40	10
2	20	20	–	–
3	20	20	20	5
4	15	15	15	5
5	10	10	20	2.5
6	10	10	10	2.5
7	5	5	10	2
8	5	5	5	1

The greatest purification effect for visual comparison of settled filtrate-reagent mixtures № 1, № 2, № 3 and №5 is shown in Fig. 10.

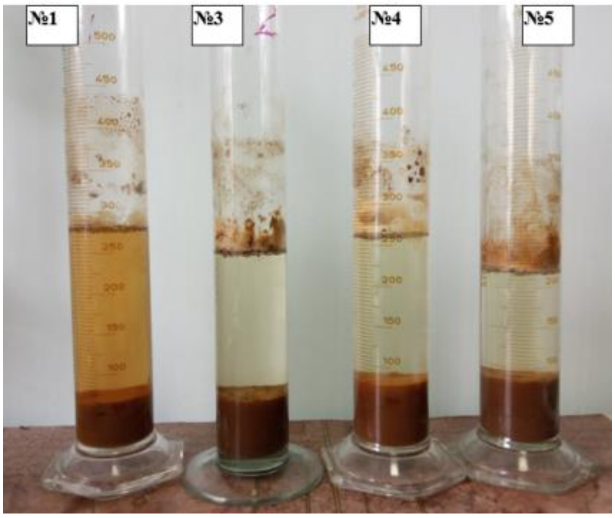

Fig. 10. Filtrate-reagent mixtures № 1, № 3, № 4 and № 5 after sedimentation

Thus, based on the obtained results of filtrates reagent purification, the doses that were used in mixtures № 3– № 5 can be considered the most optimal doses of working solutions of reagents, namely:

1) aqueous solution of PAA (0.1% wt.): In the range of 10–20 cm^3 per 200 cm^3 of aerobically purified filtrate (0.05–0.1 volume fraction);

2) aqueous solution of aluminium sulfate (10% wt.): 10–20 cm^3 per 200 cm^3 of filtrate (0.05–0.1 vol.h.);

3) aqueous solution of ferrous sulfate (II) (10 wt.%): 15-20 cm^3 per 200 cm^3 of filtrate (0.075-0.1 vol.h.);

4) aqueous solution of hydrogen peroxide (10 % wt.): 2.5–5 cm^3 per 200 cm^3 of filtrate (1.25–4.0 % vol.).

4. Conclusion

– New experimental results were obtained on the implementation conditions and aerobic biochemical filtrates purification efficiency, as well as filtrates reagent purification using the modified Fenton method.

– According to the results of laboratory studies of a typical Chervonohrad landfill filtrate (Lviv region) the optimal operating parameters of aerobic biochemical treatment stages and reagent treatment of typical Ukrainian landfills and dumps filtrates are obtained with the proposed two-stage technology.

– In laboratory conditions, the recommended intensity and duration of the filtrate pneumatic aeration at the stage of biochemical purification is defined.

– The composition and the most effective concentrations of working solutions of reagents are established experimentally.

– In laboratory conditions, time parameters of reagents working solutions introduction in the structure of reagent treatment, the intensity and duration of mixing and subsequent sedimentation of mixtures in the structure of filtrates reagent treatment are established.

– Based on the analysis of laboratory tests and comparison of the obtained results with the data of previous studies, the optimal parameters of biochemical and reagent stages of typical Ukrainian landfills and dumps filtrates treatment are established. This will allow obtaining the main indicators of pollution (BOD, COD, nitrogen, suspended solids, pH) of the treated filtrate is less than the limits of discharge into the municipal sewerage system of Ukraine according to the “Rules of acceptance of wastewater into centralized drainage systems” requirements which are approved by the Order of the Ministry of Regional Development, Construction and Housing of Ukraine from 01.12.2017 № 316, namely:

– medium reaction (pH): in the range of 6.5–9.0;

– biochemical oxygen demand (BOD): 350 mg/dm^3;

– chemical oxygen demand (COD): 500 mg/dm^3;

– nitrogen content (the sum of organic and ammonium nitrogen): 50 mg / dm^3;

– suspended solids and surfactants content: 300 mg / dm^3.

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