

# Application of two-stage aerobic-reagent technology for purifying filtrates of dumps and municipal solid waste landfills

## Zastosowanie dwustopniowej technologii tlenowo-chemicznej do oczyszczania odcieków z wysypisk i składowisk stałych odpadów komunalnych

Vronska N.Yu., Malovanyy M.S., Zhuk V.M., Tymchuk I.S., Grechanyk R.M., Soloviy Kh.M., Slyusar V.T., Marakhovska A.O.\*)

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### Abstract

Studies of the filtrate of a typical Chervonohrad municipal solid waste (MSW) landfill (Lviv region, Ukraine) were conducted and the optimum operating parameters of stages of aerobic biochemical treatment and reagent treatment of typical filtrate of Chervonohrad MSW landfill by two-stage technology were obtained. Experimental results on the conditions of implementation and efficiency of aerobic biochemical purification of filtrates, as well as reagent purification of filtrates by the modified Fenton method were obtained.

### Introduction

In Ukraine, the situation with the collection, processing and disposal of waste in general is critical and requires an urgent solution. 441.5 million tons of waste were generated in 2019, which is 20.2% more than in 2018. 98.7% (435.6 million tons) are waste generated as a result of economic activity, incl. 88.5% of the total (390.6 million tons) – in the extractive industry and quarrying, as well as 1.3% (5.9 million tons) – in households [1].

In 2019, 52.9 million m<sup>3</sup> of municipal solid waste (MSW) was collected, of which 49.7 million m<sup>3</sup> of solid waste was disposed of in landfills and dumps, which is about 94.2% of the amount of solid waste collected.

On the territory of Ukraine the area of landfills and dumps is 8838 hectares, of which the area of 1.7 thousand hectares does not meet safety standards. As of 2020, there were 6,073 landfills and dumps in Ukraine. 258 units were overloaded, and 905 units do not meet safety standards.

One of the most dangerous consequences of non-regulatory disposal of MSW for the environment is the contamination of soils, as well as surface and groundwater by leachate (or leachate). Every year, 2.2 to 5.0 million m<sup>3</sup> of filtrates are generated at landfills and MSW dumps in Ukraine. Depending on the degree of dilution by atmospheric water, the concentration of major pollutants in the filtrates is 5-50 times higher than the limit values. Highly toxic leachates enter soils, surface water and groundwater, causing unforeseen damage to the environment and human health.

Aerobic methods of biological treatment of leachates are flexible in use – they quickly adapt to the variable composition and consumption of infiltrates. Aerobic reactors, unlike anaerobic ones, are cheaper, easier to use, and much easier to operate.

In the United Kingdom, Germany, and other European countries, reagent-type leachate treatment methods such as coagulation and flocculation are used for pre – or complete treatment of MSW landfill filtrates. These methods remove biologically non-oxidizing humic and fulvic acids from the filtrates. The classical Fenton system uses iron (II) ions for the catalytic radical decomposition of hydrogen peroxide. Studies have shown

that the effectiveness of purification of filtrates from organic contaminants using traditional or modified types of Fenton process depends on the type of filtrate to be purified, dose of reagents, method of introduction and mixing, initial and final pH values, temperature, aeration intensity, etc. [2, 3, 4, 5].

**The purpose** of the study is a lab-scale verification of a two-stage aerobic-reagent technology for the pre-treatment of the typical filtrate of Chervonohrad MSW landfill which will allow discharge of pre-treated filtrates for the final treatment at the municipal wastewater treatment plants.

### Methods and techniques

The filtrate of the Chervonohrad MSW landfill (Lviv region, Ukraine) was used for laboratory tests. Drainage and filtrate collection was carried out in accordance with the requirements of regulatory documents [6, 7, 8]. The filtrate storage tank is shown in Fig. 1.



Fig. 1. Filtrate storage tank at the Chervonohrad MSW landfill  
Rys. 1 Zbiornik odcieku na składowisku MSW Chervonohrad

\*) Vronska N.Yu., C.Sc.(tech), Prof. Assoc., Lviv Polytechnic National University, Malovanyy M.S., D.Sc.(tech), prof., Lviv Polytechnic National University, myroslav.mal@gmail.com, Zhuk V.M., C.Sc.(tech), Prof. Assoc., Lviv Polytechnic National University, Tymchuk I.S. – C.Sc.(agriculture), Prof. Assoc., Lviv Polytechnic National University, Grechanyk R.M. – C.Sc.(agriculture), Prof. Assoc., Department of Ecology and Natural Resources of Lviv State Administration, Soloviy Kh.M. – PhD, Lviv Polytechnic National University, Slyusar V.T. – C.Sc.(tech), Lviv Polytechnic National University, Marakhovska A.O. – Lviv Polytechnic National University

*Aerobic biochemical purification of the filtrate.* The efficiency of pre-purification of the filtrate in the laboratory was performed on the following main indicators of contamination: reaction of the environment (pH); biochemical oxygen demand ( $BOD_{tot}$ ), chemical oxygen demand (COD), Kjeldahl nitrogen (sum of organic and ammonium nitrogen), suspended solids and floating substances (SS). In addition to the main indicators, auxiliary, technological parameters of the filtrate quality were determined at different stages of its purification, namely: biochemical oxygen consumption for the first 5 days ( $BOD_5$ ) and ammonium nitrogen concentration ( $NH^+$ ).

Laboratory studies of aerobic filtrate purification were performed according to the following method. 4 dm<sup>3</sup> of filtrate was poured into a glass laboratory flask with a capacity of 5 dm<sup>3</sup>. The filtrate purification studies were performed at a constant study temperature of  $20 \pm 1$  °C. Air with a volume flow rate of 0.05 dm<sup>3</sup>/s was supplied from the laboratory compressor to the lower part of the flask through the laboratory aerator. Aeration was carried out in a continuous mode. Air flow for aeration was measured with a laboratory rheometer and controlled by the position of the control valve. After certain periods of time, samples were taken in which the above parameters were determined.



Fig. 2. Experimental unit for the study of the efficiency of biochemical aerobic treatment of the Chervonohrad MSW landfill filtrate

Rys. 2 Stanowisko badawcze efektywności biochemicznego tlenowego oczyszczania odcieków ze składowiska odpadów komunalnych Chervonohrad MSW

*Reagent purification of filtrates.* For laboratory verification reagent purification stage of MSW landfill filtrates (the second stage of the proposed technology) a series of experimental studies was performed with the addition of aerobically purified in the first stage filtrate aqueous solutions of four reagents: polyacrylamide (PAA), aluminium sulphate, ferrous (II) sulphate and hydrogen peroxide. Before the adding to the aerobically pre-treated filtrate, working aqueous solutions of these reagents with different mass concentrations were prepared. The optimum values of concentrations of reagent working solutions were established:

- PAA: 0.1% mass solution (10–3 kg/dm<sup>3</sup> of solution or 1000 ppm);
- aluminium sulphate: 10 wt% solution;
- ferrous (II) sulphate: 10 wt% solution;
- hydrogen peroxide: 10 wt% solution.

The volume of biochemically pre-treated filtrate in all experiments was the same and equal to  $W_f = 200$  cm<sup>3</sup>. Laboratory simulation of reagent treatment was performed in laboratory glass cylinders with a volume of 500 cm<sup>3</sup>.

The aim of this study was to determine the optimum reagent compositions that give the maximum possible effect of purification by ammonium nitrogen,  $BOD_5$  and COD, the kinetics of subsequent precipitation of a mixture of filtrates with reagent solutions.

COD values were determined according to the national standard of Ukraine DSTU ISO 6060: 2003 [9]. The concentration of ammonium nitrogen was determined by the photometric method of ammonium ions

with Nesler's reagent due to the requirements of KND 211.1.4.030-95 [10]. A photoelectric colorimeter KFK-2-IHL4.2 was used. The value of  $BOD_5$  was determined by the method of analysis of the biochemical oxygen demand after n days (BOD) in natural and wastewater (KND 211.1.4.024-95) [11]. The pH value was determined using a pH meter Checker 1 HI98103 according to the relevant instruction.

## Results and discussion

After aerobic biochemical purification of the filtrate, the parameters of the purified filtrate were obtained, which are shown in table 1.

Table 1

Changes in filtrate after aerobic biochemical purification

Wskaźniki jakości odcieku przed i po biochemicznym tlenowym oczyszczaniu

Pollutant indicators	Initial filtrate parameters	Filtrate parameters after aerobic biochemical purification
pH	9.8	9.6 ÷ 10.2
Ammonium nitrogen	1427 mg/dm <sup>3</sup>	212.6 mg/dm <sup>3</sup>
Kjeldahl nitrogen	2253 mg/dm <sup>3</sup>	358.2 mg/dm <sup>3</sup>
COD	5768 mg O <sub>2</sub> /dm <sup>3</sup>	4000 mg O <sub>2</sub> /dm <sup>3</sup>
$BOD_5$	128.5 mg O <sub>2</sub> /dm <sup>3</sup>	30 mg O <sub>2</sub> /dm <sup>3</sup>
$BOD_{tot}$	507.5 mg O <sub>2</sub> /dm <sup>3</sup>	101 mg O <sub>2</sub> /dm <sup>3</sup>
Content of suspended solids and floating substances	228 mg/dm <sup>3</sup>	251 mg/dm <sup>3</sup>

In the process of biochemical aerobic purification of the filtrate, the concentration of ammonium nitrogen decreased by 6.7 times, and the concentration of nitrogen (the sum of organic and ammonium nitrogen) decreased by 6.3 times. The value of COD decreased by only 30%, so the filtrate needs further purification.

At the beginning of the experiment, there was a sharp decrease in the level of  $BOD_5$ , which is probably associated with the oxidation of easily oxidizable biological compounds. In the future, the oxidation of biological contaminants is slowed down. In general, for the entire period of the study, the value of  $BOD_5$  decreased by almost 5 times.

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

The efficiency of the Fenton reaction is higher the higher the concentration of hydrogen peroxide in the working area. Keep in mind that hydrogen peroxide is a non-flammable but explosive liquid, a strong oxidant that decomposes rapidly in open containers to water and oxygen, mixes and dissolves in any ratio with water. Therefore, the concentration of the working solution of hydrogen peroxide was set primarily based on the conditions of safe implementation of the process in the laboratory, as well as in research and industrial and in-kind industrial conditions.

As a result of laboratory studies it was found that the maximum allowable concentration of ferrous sulfate in its working solution is 10% of the mass. in terms of technical product, which corresponds to 5.3% of the mass. in terms of anhydrous ferrous sulfate (II).

Optimum doses of working reagent solutions were determined in the laboratory using pre-biochemically purified filtrate. The criterion for the optimum composition of the working solutions were the initial values of the main indicators of filtrate contamination after reagent treatment.

The composition of the reagent compositions in laboratory studies of reagent purification of filtrates are given in table 2.

All reagent compositions contained all 4 reagents, except composition №2. For this composition (№2), fast and sufficiently deep

clarification of the filtrate sample was obtained before the introduction of ferrous sulphate (II) and hydrogen peroxide. However, the low efficiency of using an incomplete set of reagents in terms of reducing COD, and contents of ammonium nitrogen and Kjeldahl nitrogen was obtained.

Table 2

The composition of the reagent compositions in the laboratory study of reagent purification of filtrates (volume of filtrate  $W_f = 0.2$  L)

Skład odczynników wykorzystanych w badaniu laboratoryjnym oczyszczania odcieków (objętość filtratu  $W_f = 0,2$  L)

No of mixture	Volumes of aqueous reagent solutions, cm <sup>3</sup>			
	PAA (0.1 wt%)	Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> ×18 H <sub>2</sub> O (10 wt%)	FeSO <sub>4</sub> ×7 H <sub>2</sub> O (10 wt%)	H <sub>2</sub> O <sub>2</sub> (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 results of laboratory determination of the main indicators of pollution after the introduction of aerobically purified filtrate of the appropriate reagent compositions and settling of the resulting mixtures are given in table 3.

Table 3

Filtrate pollutant indicators after the reagent purification step

Wskaźniki jakości oczyszczonych odcieków po etapie oczyszczania chemicznego

Pollutant indicators, units of measurement	Mixture number								
	№ 0*	№ 1	№ 2	№ 3	№ 4	№ 5	№ 6	№ 7	№ 8
pH	10	4.8	7.6	6.6	6.8	6.7	7.2	7.6	8.5
Ammonium nitrogen, mg/dm <sup>3</sup>	212.7	5.6	35.3	10.2	8.8	9.6	17.8	14.2	15.5
Kjeldahl nitrogen, mg/dm <sup>3</sup>	358	9.9	61.2	17.3	15	16.3	30.3	24.2	26.4
BOD <sub>5</sub> , mg O <sub>2</sub> /dm <sup>3</sup>	26	9.5	18.8	15.9	13.5	17.1	14.3	18.6	20.8
BOD <sub>tot</sub> , mg O <sub>2</sub> /dm <sup>3</sup>	102.7	39.5	75	62.3	54.8	68.4	57.2	74.4	83.2
COD, mg O <sub>2</sub> /dm <sup>3</sup>	4038	123.6	1195	412	432	394	489	537	625
Suspended solids and floating substances, mg/dm <sup>3</sup>	251	84.5	58.7	34.3	62	71.4	57.8	124.8	145.2

\* filtrate after the aerobic purification stage

The highest effects of purification of the settled filtrate-reagent mixtures were obtained for the samples №1, №2, №3 and №5. Thus, based on the results of reagent purification of filtrates, it was found that the optimum dosage of working reagent solutions corresponds to mixtures №3– №5, namely:

- 1) aqueous solution of PAA (0.1 wt%): in the range of 10–20 cm<sup>3</sup> per 200 cm<sup>3</sup> of aerobically purified filtrate (5–10 vol.%);
- 2) aqueous solution of aluminium sulphate (10 wt%): 10–20 cm<sup>3</sup> per 200 cm<sup>3</sup> of filtrate (5–10 vol.%);
- 3) aqueous solution of ferrous (II) sulphate (10 wt%): 15–20 cm<sup>3</sup> per 200 cm<sup>3</sup> of filtrate (7.5–10 vol.%);
- 4) aqueous solution of hydrogen peroxide (10 wt%): 2.5–5 cm<sup>3</sup> per 200 cm<sup>3</sup> of filtrate (1.25–2.5 vol.%).

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## Conclusions

New experimental results on the parameters of realization and efficiency of aerobic biochemical filtrate purification followed by reagent purification using the modified Fenton's method are obtained in the lab-scale conditions for the filtrate of the Chervonohrad MSW landfill (Lviv region, Ukraine).

High efficiency of the aerobic purification of the Chervonohrad MSW landfill filtrate is obtained by ammonium nitrogen, Kjeldahl nitrogen and BOD.

The composition and the most effective concentrations of working solutions of reagents are determined. Optimal dosage of aqueous working solutions for the filtrate of the Chervonohrad MSW landfill are as follows PAA (0.1 wt%): 5–10 vol%, aluminium sulphate (10 wt%): 5–10 vol%, ferrous (II) sulphate (10 wt%): 7.5–10 vol% and hydrogen peroxide (10 wt%): 1.25–2.5 vol.%.

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