

## Integrated Aerobic-Reagent Technology for the Pre-Treatment of Leachates from Municipal Solid Waste Landfills

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### ABSTRACT

Accumulation of leachate at municipal solid waste (MSW) landfills is a significant environmental problem. The analysis of known technologies of leachate treatment was performed. It was established that it is not effective to use the same technology to treat leachate in two different periods: before the landfill closure and reclamation process as well as afterwards. The application of integrated two-stage aerobic-reagent pre-treatment technology with subsequent full treatment at municipal wastewater treatment plants was proposed for these purposes. The results of laboratory studies of optimal parameters of technology realization for the pre-treatment of Lviv MSW landfill leachate were presented. Recommendations for the practical implementation of the combined two-stage aerobic-reagent technology for the landfill leachate pre-treatment were developed.

**Keywords:** aerobic-reagent pre-treatment, landfill, leachate, modified Fenton process, municipal solid waste.

### INTRODUCTION

According to the report of the International Finance Corporation (World Bank Group) (Municipal Solid Waste in Ukraine, 2015), 11–13 million tons of municipal solid waste (MSW) are generated in Ukraine annually, and more than 90% are stored at landfills and dumps. The total number of waste storage facilities is about 6700, and their total area is estimated as 10,000 hectares. On the basis of the average annual height of the precipitation layer of 500 mm/year and the values of the runoff coefficient of 0.05–0.1, overall about 2.5–5 million m<sup>3</sup> of leachate are formed annually at these facilities. The largest volumes of leachate (100,000 m<sup>3</sup> and above) are collected in storage ponds at the largest Ukrainian landfills,

in particular, at the Kyiv and Lviv landfills. Depending on the degree of dilution by atmospheric water, the concentrations of the main pollutants in leachates are 5–50 times higher than the limiting values. Due to the lack of organized collection and treatment of leachate at many landfills and dumps, highly toxic leachate leaks directly into soils, underground and surface water bodies, causing great damage to the environment.

The problems of the landfill leachate treatment are relevant during the design, operation and planned closure of these facilities. At the design stage, as a rule, an innovative technology of treatment of the leachate collected by the drainage system is laid, the productivity of which corresponds to the calculated one. During operation, the priorities in choosing the method of treatment

of leachate depend on the history of operation and the state of the leachate drainage system. It is necessary to distinguish between the systems of landfill leachate treatment that meet the requirements of DBN V.2.4-2-2005 and Directive 1999/31/EC, and in the planned mode operate the designed and installed units, as well as the systems of landfill leachate treatment, which are extremely rare, and the existing ones work inefficiently and do not provide the required degree of treatment.

In solving the problem of eliminating the environmental danger caused by the leachate from landfills and dumps in Ukraine, it is necessary to distinguish two characteristic periods at the stage of their closure and reclamation:

- period #1: short-time treatment of high discharges of accumulated leachate in order to facilitate the rehabilitation of the landfill or dump;
- period #2: continuous treatment of low discharges of leachate, which will permanently, for decades, continue to be formed in the body of a closed landfill or dump as a result of biological decomposition processes of the organic waste component.

Since these two cases differ significantly in the volume discharge and composition of leachate as well as in the duration of implementation, it is not efficient to implement a single technology at these two stages, both from a technical point of view (inability to ensure full load and efficient operation of equipment in the second period) and from a financial point of view.

The biological aerobic methods are the most common for urban wastewater treatment (Iurchenko et al., 2019; Iurchenko et al., 2020), although a lot of studies were performed on the use of anaerobic (Malovanyy et al., 2014; Sabliy et al., 2019) and adsorption (Malovanyy et al., 2019a; Malovanyy et al., 2019b) technologies. Adsorbents synthesized from organic waste (Shmandy et al., 2012), inorganic waste (Zasidko et al., 2019), vegetable raw materials (Shmandiy et al., 2020), natural sorbents (Bezdeneznych et al., 2020) are often used in the adsorption technologies. The technologies for treatment of leachate using bioplateau are of special interest (Popovych et al., 2018; Popovych et al., 2020). Insufficiently treated wastewater enters the surface water and causes eutrophication (Nykyforov et al., 2016).

Up to date the following technologies to treat the landfill leachate have been the most common (DBN V.2.4-2-2005; Gao et al., 2015):

- 1) reverse osmosis technology;
- 2) evaporation and drying technology;
- 3) leachate binding technology;
- 4) technologies of anaerobic and aerobic biochemical treatment.

Technologies 1–3 are energy and resource-consuming, their implementation requires significant construction and operation costs; thus, their practical realization is often inefficient. The technology of anaerobic leachate treatment can be potentially effective, but for its successful implementation on an industrial scale it is necessary to strictly adhere to the operating parameters, which is difficult to do in practice under the conditions of variable quality composition of the leachate. Different schemes of leachate treatment in aerated lagoons, often combined with anaerobic ponds and with constructed wetlands, were successfully tested in Great Britain, Norway, Sweden and other countries (Mæhlum, 1995; Mehmood et al., 2009; Robinson & Grantham, 1988). Effective leachate treatment was obtained combining aerated lagoon with the upflow anaerobic sludge blanket reactor (Govahi et al., 2012).

The reagent methods, in particular coagulation and flocculation, chemical oxidation and Fenton process are widely used for pre-treatment or complete treatment of MSW landfill leachate in Germany, Great Britain and other countries. These methods allow removing from the leachate biologically non-oxidizing humic and fulvic acids, as well as other specific heavy contaminants, including heavy metal ions, organochlorine compounds etc. It was obtained that the Fenton process is effective for the treatment of mature leachate with especially high content of organics, e.g. maximum effects 77% and 83% of COD reduction were obtained (Badawy et al., 2013) for initial COD values 6250 and 13300 g/m<sup>3</sup>, respectively.

Combined methods of leachate treatment are of the special interest due to different possible synergetic effects, depending of the leachate composition. High efficiency of the leachate treatment was obtained combining the Fenton process and aerobic activated sludge process (Bae et al., 1997). The leachate treatment plants in Germany, as usual, use combined treatment methods; at more than 60% of such plants the first stage is biochemical treatment, and 15 plants use the chemical oxidation processes for the subsequent treatment stage.

The previous analysis indicated that the two-stage combined technology of leachate pre-treatment, including biochemical stage in aerated lagoons followed by reagent treatment using the modified Fenton process could be especially effective for the implementation at Ukrainian landfills and dumps (Malovanyy et al., 2018).

## MATERIALS AND METHODS

The leachate of the Lviv MSW landfill was used for research. Main input parameters of the raw leachate: pH – 8.64; concentration of ammonium ions – 900 g/m<sup>3</sup>; chemical oxygen demand (COD) – 11000 g/m<sup>3</sup>.

Laboratory modelling of the first (aerobic) stage of the leachate pre-treatment was performed on an experimental unit in a flask with a capacity of 5 dm<sup>3</sup> as small-scale model of the aerated lagoon. The study of aerobic leachate pre-treatment was performed at a constant temperature of 20 °C. A laboratory aerator was installed at the bottom of the flask, and laboratory compressor supplied air at a constant flow rate of 42 cm<sup>3</sup>/s. Aeration was performed in a continuous mode. The samples were periodically taken from the flask to measure the COD value and the content of ammonium ions. The value of pH and the concentration of dissolved oxygen in the treated medium were checked regularly.

At the second (reagent) stage of the pre-treatment, different modifications of the Fenton's reagent composition were investigated to achieve the maximum treatment effect at the minimum possible costs of the composition. In the testing process, the proportions of the reagent were changed, the effects of mixing and aeration and the effect of adding the polyacrylamide (PAA) solution as a flocculant were investigated (Table 1). Final content of contaminants was the main criterion of the effectiveness of the reagent composition.

## RESULTS AND DISCUSSION

### Laboratory investigation of the optimum parameters of the aerobic-reagent technology application for Lviv MSW leachate pre-treatment

Analysis of the results of aerobic biochemical treatment studies showed that the maximum effect of leachate treatment by COD and ammonium ion concentration under selected study conditions is achieved asymptotically after 16 days from the beginning of the process.

The microbiological analysis revealed the appearance in the leachate of a wide range of microbiological aerobic culture, which differs significantly from the culture of activated sludge of municipal wastewater treatment plants (WWTP). For all investigated values of hydraulic retention time (HRT) of the leachate in the laboratory model of the aerated lagoon, the concentration of dissolved oxygen in the test medium was maintained at 3.9 mg/dm<sup>3</sup>. On average, the steady mode of unit operation in dynamic mode was reached after 9–11 days. The optimum time of hydraulic retention of the leachate in the lagoon in the implementation of biological aerobic treatment for the selected study conditions is a period of 11 days. In this case, the effect of treatment of leachate from ammonium ions by 35% and reduction of COD by 50% was achieved. However, for two-stage technology, the optimum duration of the first stage is determined on the basis of technical and economic assessment of integrated technology as a whole for each case of practical implementation of the process.

In the process of laboratory modelling of the reagent treatment stage of the leachate, different types of reagents were gradually added in the investigated fresh leachate in order to separate the phases by coagulation, flocculation and flotation of contaminants. The study was performed in measuring

**Table 1.** Investigated reagent compositions for the Lviv MSW landfill leachate treatment on the reagent stage

| N | Reagent dosage, kg/m <sup>3</sup>    |   |                                      | Other options |          |
|---|--------------------------------------|---|--------------------------------------|---------------|----------|
|   | FeSO <sub>4</sub> ·7H <sub>2</sub> O | Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> ·18H <sub>2</sub> O | H <sub>2</sub> O <sub>2</sub> (60 %) | PAA           | aeration |
| 1 | 3.2                                  | 1.9   | 1.6                                  | -             | -        |
| 2 | 2.8                                  | 1.0   | 0.51                                 | -             | -        |
| 3 | 3.2                                  | -   | 1.06                                 | +             | -        |
| 4 | 3.2                                  | 1.9   | 1.6                                  | +             | -        |
| 5 | 2.8                                  | 1.0   | 0.51                                 | +             | -        |
| 6 | 2.4                                  | 0.9   | 0.5                                  | +             | +        |
| 7 | 2.1                                  | 0.7   | 0.45                                 | +             | +        |

cylinders. Reagents or reagent compositions were added portionwise to a 0.2 dm<sup>3</sup> aliquot of leachate. Different variants of mixing and aeration of the investigated medium after dosing of reagents were investigated. The control of the reagent treatment process of the leachate was performed by changing the optical density of the medium. The efficiency of treatment by using the reagent method was judged by the change in the concentrations of contaminants in the test medium. As the results of the research showed, the best results were obtained when using the modified Fenton’s method. Probably, a high treatment effect is achieved due to the oxidation of suspended and dissolved components with hydrogen peroxide; in turn, iron salts, as a component of the Fenton’s reagent, more effectively perform the role of coagulant. Various variants of modifying the composition of Fenton’s reagent in order to achieve the maximum degree of treatment and the minimum cost of the reagent composition were investigated. In the process of studying these options, the dosage of the reagent composition was adjusted; the effects of duration and intensity of mixing and aeration of the studied system and the effect of adding the flocculant were studied.

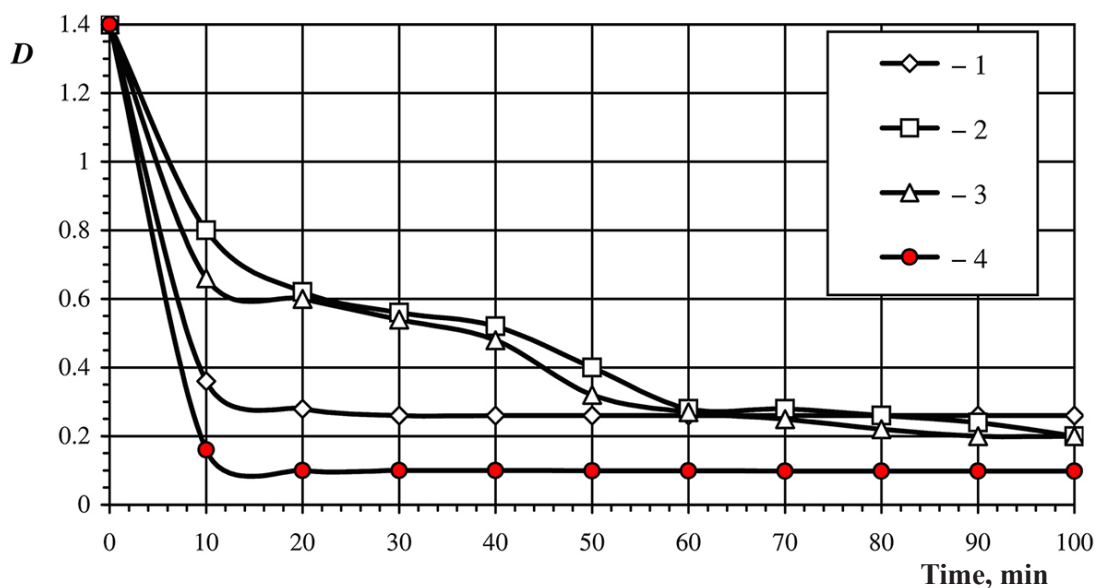
At the first step of the study of the leachate reagent pre-treatment, the most effective reagents, which ensure the maximum efficiency of the contaminants separation from the leachate, were selected. The reagents which are most commonly used in the industrial wastewater treatment were investigated for this purpose: hydrates of aluminium sulphate

and iron (II) sulphate, aluminium polyoxochloride, as well as Fenton’s reagent. The kinetics of the optical density changing was measured after adding the reagent solutions into the leachate. The best results were obtained in the case of using the Fenton’s reagent (Fig. 1). This result can be explained by intensive oxidation of suspended and dissolved components of leachate by hydrogen peroxide. The ferrous salts that are the part of the Fenton’s reagent also act as a coagulant in this case.

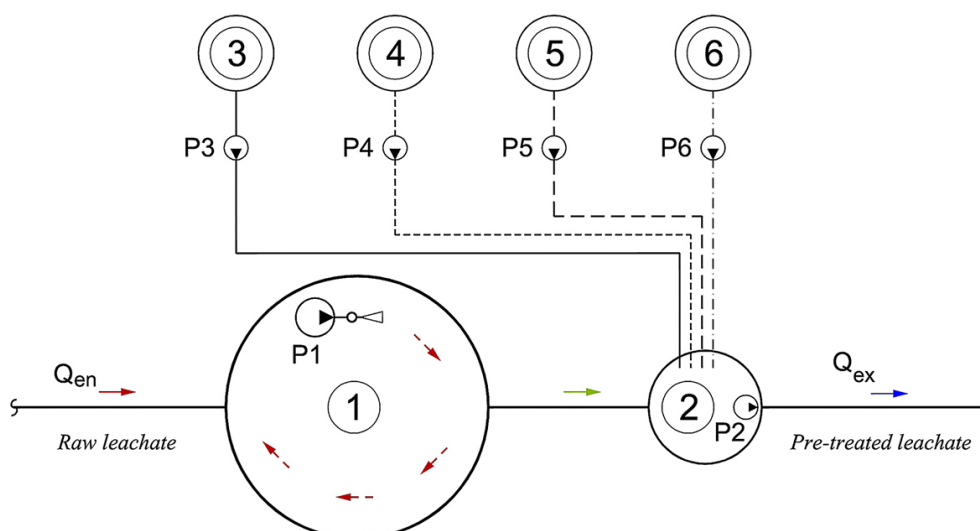
It was found that none of the reagent composition allows obtaining the limiting concentration for discharge into the municipal sewerage system after pre-treatment by COD parameter. A positive result was obtained only when using the integrated two-stage technology of treatment of leachate by sequential implementation of aerobic biochemical treatment for a sufficiently long HRT and reagent treatment of the leachate by the modified Fenton method.

### Recommendations for the practical implementation of aerobic reagent technology for pre-treatment of solid waste landfill leachate

On the basis of the analysis of the results of laboratory research, the scientific and practical recommendations for the introduction of energy-efficient and resource-saving two-stage technology for the pre-treatment of leachates at landfills and dumps in Ukraine were developed.



**Figure 1.** Lviv MSW landfill leachate optical density changing using the different reagents: 1 –  $Al_2(SO_4)_3 \cdot 18H_2O$ ; 2 –  $FeSO_4 \cdot 7H_2O$ ; 3 –  $Al_n(OH)_{(3n-m)}Cl_m$ ; 4 – Fenton’s reagent



**Figure 2.** Scheme of prototype unit for the leachate pre-treatment at MSW landfills and dumps: 1 – biochemical pre-treatment reactor; 2 – reagent pre-treatment reactor; 3–6 tanks with solutions of reagents, respectively  $\text{Al}_2(\text{SO}_4)_3$ ,  $\text{FeSO}_4$ ,  $\text{H}_2\text{O}_2$ , PAA; P1– jet aerator pump; P2– sludge pump; P3 – P6 – centrifugal pumps

The two-stage technology consists in the sequential implementation of the stage of aerobic biochemical treatment and the stage of reagent treatment of the leachate, which take place in two separate reactors (Fig. 2).

The biochemical treatment stage should be realized in the continuous reactor for a sufficiently long time (from 1 day to 30 days) and consists in aeration of the medium by means of a jet-type aerator pumps and, accordingly, in deep aerobic biochemical treatment of the leachate. The reagent treatment stage should take place in a batch reactor. Small portions of the leachate after the first stage of treatment enter the reagent reactor, after which there are pressurized with certain volumetric discharges of solutions of four reagents: aluminium sulphate (III), iron sulphate (II), hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) and polyacrylamide (PAA). The resulting mixture is stirred vigorously, and then settled for about 30–60 minutes with the formation in the middle part pre-treated leachate, in the lower part – sludge and in the upper part – float sludge. Thus, a new type of modified Fenton's method is implemented in the reagent treatment of leachate.

The leachate pre-treated by proposed two-stage technology in a mixture with domestic and industrial wastewater should be pumped for full treatment at the municipal WWTP. For this purpose, as a result of preliminary treatment of the leachate according to the proposed two-stage technology, the concentrations of basic pollutants will be achieved not higher than the relevant values of Ukrainian discharge limits in sewerage systems,

namely: pH value – within 6.5–9.0; biochemical oxygen demand ( $\text{BOD}_{20}$ ) –  $350 \text{ mg/dm}^3$ ; COD –  $500 \text{ mg/dm}^3$ ; nitrogen content (sum of organic and ammonium nitrogen) –  $50 \text{ mg/dm}^3$ ; suspended solids –  $300 \text{ mg/dm}^3$ . Thus pre-treated leachate after the obtaining of the necessary discharge permission for the output into the municipal sewerage system can be discharged for full treatment with a mixture of domestic and industrial wastewaters to municipal WWTP. The sludge generated during the operation of the two-stage pre-treatment of leachates can be pumped by a sludge pump into specially prepared cells on the territory of the landfill for final disposal. It is possible also to use the sludge in the mix with natural soil to cover the solid waste at landfill [Martínez-Cruz et al., 2021].

The main practical task of integrating the technologies at the level of prototype treatment unit is to harmonize the parameters of the implementation of both stages of pre-treatment in order to achieve the desired overall efficiency of leachate treatment at a minimum total costs. Thus, at the first stage, it is planned to investigate the optimum intensity and frequency of aeration, as well as the input of special additives to adjust the ratio of carbon, nitrogen and phosphorus to optimize the biochemical treatment process. At the stage of reagent treatment, the important issues for testing are the optimum concentrations and volume discharges of reagents, the sequence and time intervals between the input of different reagents, intensity and duration of mixing and optimal duration of settling.

## CONCLUSIONS

It was established that significant threat to the sustainable development of modern cities is caused by negative impacts of the MSW landfills, in particular due to the pollution of the environment by the landfill leachate. The analysis of the leachate treatment technologies and the results of the performed experimental studies verify the prospect of application of the integrated two-stage technology, including biochemical pre-treatment under aerobic conditions followed by reagent pre-treatment using the modified Fenton's process. The optimum parameters of the implementation of both stages of leachate pre-treatment in lab scale were found. Recommendations for the practical implementation of the combined two-stage aerobic-reagent technology for the landfill leachate pre-treatment were developed.

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