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Optimal parameters for reagent treatment of Hrybovychi landfill leachates at the pilot-scale treatment plant

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Abstract. Promising technologies for the treatment of solid waste landfill leachates are considered with an emphasis on reagent and combined methods. The purpose of this study was to estimate at the pilot-scale level optimal technological parameters of the leachate oxidation using the modified Fenton process, accompanied by the simultaneous coagulation-flocculation followed by gravitational sedimentation. Pilot-scale leachate treatment plant was installed at the Hrybovychi MSW landfill (Ukraine), and reagent treatment of leachate was a second stage treatment after the aerobic biological pre-treatment. Reagent treatment unit worked in a batch mode, with nominal volume of treated leachate 100 dm^3 per cycle. Dependencies of the key pollution indicators (ammonium nitrogen, total Kjeldahl nitrogen, BOD, COD, pH, suspended solids) versus the dosage of reagent solutions are obtained. The optimum specific dosages of reagent solutions are found to be equal: 0.04 m^3 of PAA 0.1 wt% solution per 1 m^3 of leachate; $Al_2(SO_4)_3 \times 18\ H_2O$ (10 wt.%) – 0.03–0.04 m^3/m^3; $FeSO_4 \times 7H_2O$ (10 wt.%) – 0.06–0.08 m^3/m^3; hydrogen peroxide (10 wt.%) – 0.04–0.05 m^3/m^3. High efficiency of COD reduction (88.2–89.5%) is obtained at optimal doses of reagent solutions, and the optimum $[H_2O_2]$/COD ratio was found to be 0.23–0.25. Obtained maximum effects of COD reduction significantly exceed corresponding effects for the simple Fenton process reported before. This result could be explained by the synergistic effect of additional flocculation and coagulation immediately before the input of Fenton reagent. Results of the study showed the efficiency of the proposed treatment technology and allow recommending this technology for the implementation at landfill leachate local treatment plants.

Keywords: landfill leachate, modified Fenton method, pilot-scale treatment plant, reagent treatment, solid waste landfill.

1. Introduction

The consequence of the strong economic and population growth in the world is the accumulation of municipal solid waste (MSW) in countries with no system to manage this type of waste. Since household waste may include nanotechnology products and medicines, this is an additional negative factor (Vambol et al., 2017; Husain Khan A. et al., 2020). The construction of landfills (which are technical structures that prevent negative impacts on the environment) and the disposal of waste dumps (which pose a major threat to the environment) are elements to prevent the negative impact of accumulated MSW on the environment (Vambol et al., 2016). During the life cycle in the localization of MSW (landfills and dumps), the biological decomposition of the organic fraction produces a significant amount of biogas (Voytovych et al., 2020) and liquid biodegradation products (Popovych et al., 2020), which are characterized by a high content of toxic organic and inorganic substances. The concentration of these pollutants is hundreds of times higher than the limit value (Urbanas et al., 2016). These liquid effluents, called leachates, are filtered through the waste and diluted by the atmosphere and groundwater and are highly concentrated hazardous pollutants in groundwater and surface waters. To control these pollutants in the hydrosphere, it is necessary to introduce an effective monitoring system (Odnorih et al., 2020) and to remove pollutants from the leachate by the introduction of technologies for waste water treatment (Iurchenko et al., 2016, Malovanyy et al., 2018) and by the introduction of energy-efficient treatment systems (Shchur et al., 2021).

A characteristic feature of landfill leachates is high content of a large number of organometallic complex compounds, especially the products of the interaction of heavy polyvalent metals with amino or humic acids. The latter is practically not subject to conventional treatment methods since they do not precipitate due to their surface-active properties and high stability and are not destroyed by oxidizing agents. Heavy metals pose a particular threat as carriers of mutagenic, embryotoxic, and carcinogenic properties, as they can accumulate in living organisms (Urbanas et al., 2016).

The most promising for the leachate treatment are combined treatment methods. The best-known examples of the use of combinations of leachate treatment methods are biological and membrane processes (Raghab et al., 2013); biological and oxidative processes (Petruk et al., 2016); biological treatment with final treatment in municipal wastewater treatment plants (Malovanyy et al., 2018) or on a bioplateau (Malovanyy et al., 2021); biological and reagent treatment (Malovanyy et al. al., 2022); and other combinations.

Purely biological methods are most often used in combination with the stages of denitrification and nitrification (Iurchenko et al., 2020), treatment by membrane processes (Dushkyn et al., 2011), and adsorption on activated carbon or natural sorbents (Petruk et al., 2016, Sakalova et al., 2019). Treatment using sorbents of natural or synthetic origin in one stage is also used in technologies for the adsorption of heavy metals (Malovanyy et al., 2019, Danchenko et al., 2017). Physicochemical methods such as coagulation-flotation, filtration, ultrafiltration, adsorption, reverse osmosis, concentrated evaporation, etc., in turn, are used successively in various combinations in the treatment of leachates (Bae et al., 1997). As a rule, these methods are associated with considerable costs for reagents and/or high energy consumption and do not allow the leachate to be treated to the desired degree so that their use is also only useful in combination and conjunction with other approaches.

The use of reagent methods is promising. The stand-alone application of this approach requires many reagents, so it is often used in combination with other methods, usually biological (Malovanyy et al., 2022). Among several reagent methods for leachate treatment, one of the most effective is the Fenton method or modifications of this method (Deng et al., 2006; Badawy et al., 2013). Recently, much attention has been paid to studying the modified Fenton method (electro-Fenton method, photo-Fenton method). It has been shown that such modifications of the Fenton method are most promising for leachate treatment (Deng, 2007; Badawy et al., 2013). In the traditional or modified Fenton process used for leachate treatment, the efficiency of such treatment depends on some factors, namely on the type and dosage of reagents, mode of their input to the leachate, physical and chemical properties of the leachate, parameters of the treatment process conditions including pH, temperature etc.

Most of the previous studies of the leachate treatment by the Fenton method were performed in the laboratory conditions, which differs significantly by the scale of flow rates and by margin conditions of the process from the full-scale design parameters. The purpose of this study is to estimate optimal technological parameters of the leachate oxidation using the modified Fenton process, enhanced by the simultaneous coagulation and flocculation, at the pilot-scale treatment unit, which allows more realistic modeling of the processes of oxidation, flocculation, coagulation and precipitation of leachate-reagent mixtures in semi-industrial conditions for the corresponding flow rates of leachate using real technological equipment.

2. Materials and methods of research

Reagent processing was studied using the leachate, sampled from the leachate storage pond at the Hrybovychi MSW landfill (Lviv region, Ukraine). Reagent treatment of leachate was considered not as a separate technology, but as a second stage treatment after the aerobic biological preliminary treatment. Key pollution parameters of this aerobically pre-treated leachate are presented in Table 1.

Table 1. Pollution indicators of aerobically pre-treated Hrybovychi MSW landfill leachate at the inflow to reagent treatment unit.

Indicator of pollution and unit of measurement	Range
pH	9.71–9.79
Ammonium nitrogen, mg/dm^3	168.8–186.6
Total Kjeldahl nitrogen, mg/dm^3	359.8 – 397.2
BOD$_5$, mg/dm^3	49.3 – 70.4
BOD$_{tot}$, mg/dm^3	191.3–217.6
COD, mg/dm^3	3599 – 3897
Suspended solids, mg/dm^3	219–225.2

This study was carried out for eight variants of the parameters of the reagent treatment process. The input parameters of the leachate obtained after the biological aerobic treatment for reagent treatment varied within limits given in Table 1 to obtain accurate data for the analysis of the effectiveness of reagent treatment of leachates, the corresponding treatment effects E were found for each controlled indicator:

$$E = \frac{C_{en} - C_{ex}}{C_{en}} \times 100\%, \tag{1}$$

where C_{en}, C_{ex} – the initial concentration before the reagent treatment and the final concentration of the contaminants in leachate, respectively.

Study was conducted at the pilot-scale treatment plant, installed in the production pavilion of the Hrybovychi MSW landfill (Fig. 1). Reagent treatment unit worked in a batch mode, with nominal volume of treated leachate 100 dm^3 per cycle.

The investigated leachate with volume of 100 dm^3 was fed from the biological aerobic treatment unit 1 through the line 7 into the reagent treatment reactor 2 (Fig. 1). Using the dosing pumps 8, working solutions of polyacrylamide (PAA) in the form of polyacrylamide-co-diallyldimethylammonium chloride, aluminium sulphate ($Al_2(SO_4)_3$) and further, simultaneously ferrous sulphate ($FeSO_4$) and hydrogen peroxide (H_2O_2) were successively fed from the tanks 3–6 into the reagent treatment reactor 2, accompanied by intensive stirring of the liquid mixture. After the end of the feeding of working solutions, the stirring was stopped, and the entire volume of the reactor was flocculated with the reaction products. The content of the reactor was precipitated for one day, and after that samples were then taken from reactor 2 to analyze the concentration of residual contaminants.

Working solutions of PAA, aluminium sulphate, and ferrous sulphate were dissolved once a day. A working solution of hydrogen peroxide was dissolved immediately before input of this solution into the reagent reactor at each study cycle. The concentrations and dosages of the working solutions tested at the pilot-scale treatment plant are given in the Table 2. Table 2 also shows the mass concentrations of the active substances in the solutions, where the mass fractions of solutions of aluminium sulphate and iron (II) sulphate are given in the terms of technical products – crystalline hydrates, and the mass concentrations of aluminium sulphate and iron (II) sulphate in the reagent reactor are given for aluminium ion and iron (II) ion, respectively. The final values of pH_{ex} in the treated leachate after reagent treatment and phase separation are also given.

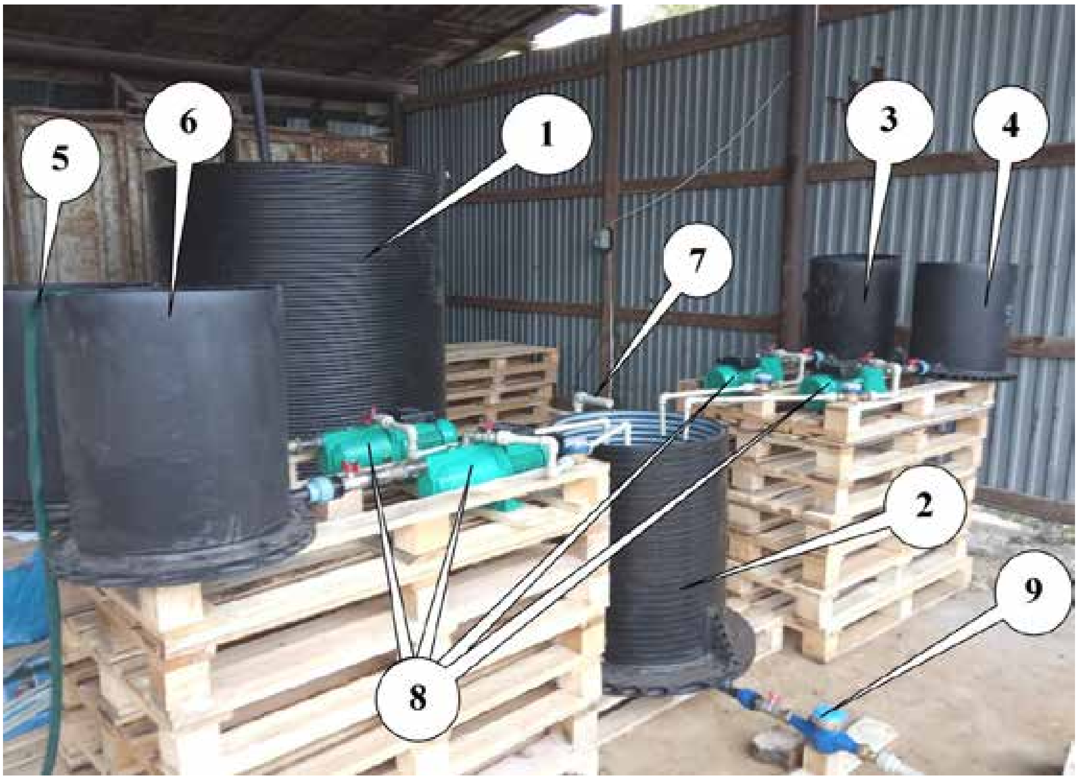

Figure 1. Experimental pilot-scale treatment plant for leachate treatment: 1 – biological aerobic treatment unit; 2 – reagent treatment reactor; 3 – tank with PAA solution; 4 – tank with solution of $Al_2(SO_4)_3$; 5 – tank with solution of $FeSO_4$; 6 – tank with hydrogen peroxide solution; 7 – feed line of aerobically pre-treated leachate into the reagent treatment reactor; 8 – dosing pumps for feeding reagent solutions into the reagent treatment reactor; 9 – water meter for accounting the treated leachate.

Table 2. Concentrations and dosages of reagent solutions and the final pH in the pilot-scale study of leachate reagent treatment at Hrybovychi MSW landfill.

Mixture	PAA (0.1 wt%)		$Al_2(SO_4)_3 \times 18\ H_2O$ (10 wt%)*		$FeSO_4 \times 7\ H_2O$ (10 wt%)*		Hydrogen peroxide (10 wt%)		pH_{ex}
	dosage, dm^3	*C*, g/m^3	dosage, dm^3	*C*,** g/m^3	dosage, dm^3	*C*,** g/m^3	dosage, dm^3	*C*, g/m^3	
No. 1	5	50	5	405.4	10	2014.4	6	1200	6.26
No. 2	5	50	4	324.3	8	1611.5	5	1000	6.63
No. 3	4	40	5	405.4	8	1611.5	5	1000	6.54
No. 4	4	40	4	324.3	6	1208.6	5	1000	6.78
No. 5	4	40	3	243.2	8	1611.5	4	800	6.67
No. 6	3	30	4	324.3	8	1611.5	4	800	6.57
No. 7	3	30	5	405.4	6	1208.6	3	600	6.7
No. 8	3	30	4	324.3	6	1208.6	3	600	6.81

Note:
* – the mass proportions of solutions of aluminium sulphate and ferrous sulphate are given in terms of marketable products;
** – the mass concentrations of aluminium and iron (II) ions are presented.

3. Results and discussion

Histograms of changes in the main indicators of leachate contamination in reagent treatment and the corresponding treatment effects are presented in Figure 2–7.

Changes of ammonium nitrogen and total Kjeldahl nitrogen (TKN) content are presented in Figure 2–3. For reagent mixtures No.1–7 TKN in the treated leachate was below the limit for discharge into Ukrainian sewerage systems (50 mg/dm^3), and only for reagent mixture No.8 slightly exceeded the limit standard – 56.2 mg/dm^3 (Fig. 3).

The most problematic indicator of pollution in terms of compliance with the sewerage discharge limits is COD, and for the mixtures No.6, No.7 and No.8 discharge limit (500 mg/dm^3) was exceeded (Fig. 4). Optimum chemical dosages are obtained for the mixtures No.4 and No.5. The use of additional flocculation and coagulation immediately before the addition of Fenton reagent allowed to obtain for reagent mixtures No.4–No.5 a high efficiency of COD reduction by 88.2–89.5% at a molar concentration of hydrogen peroxide 24–30 mmol/dm^3 and 22–29 mmol/dm^3 of iron (II) ions. Obtained maximum effects of COD

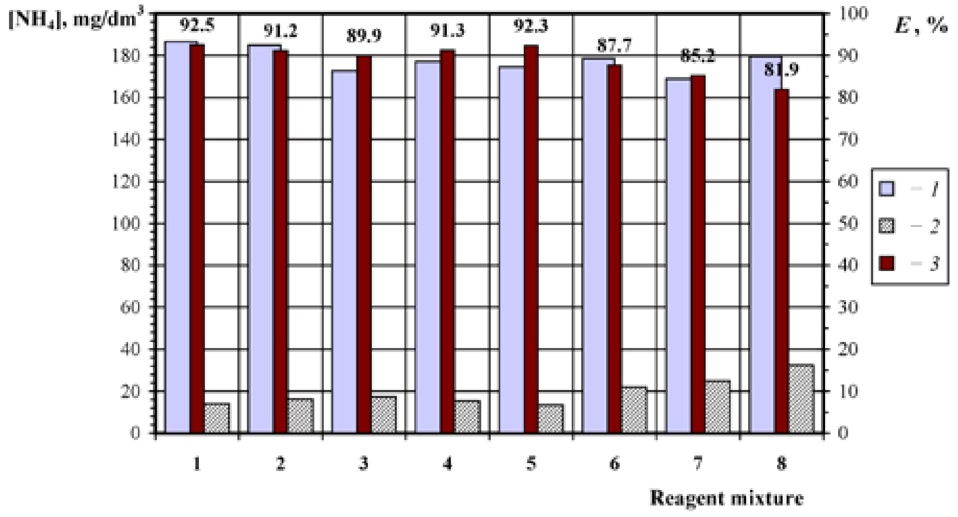

Figure 2. Results of the reagent treatment of Hrybovychi MSW leachate at the pilot-scale treatment plant by the ammonium nitrogen: 1 – before the reagent treatment; 2 – after the reagent treatment; 3 – treatment effect, %.

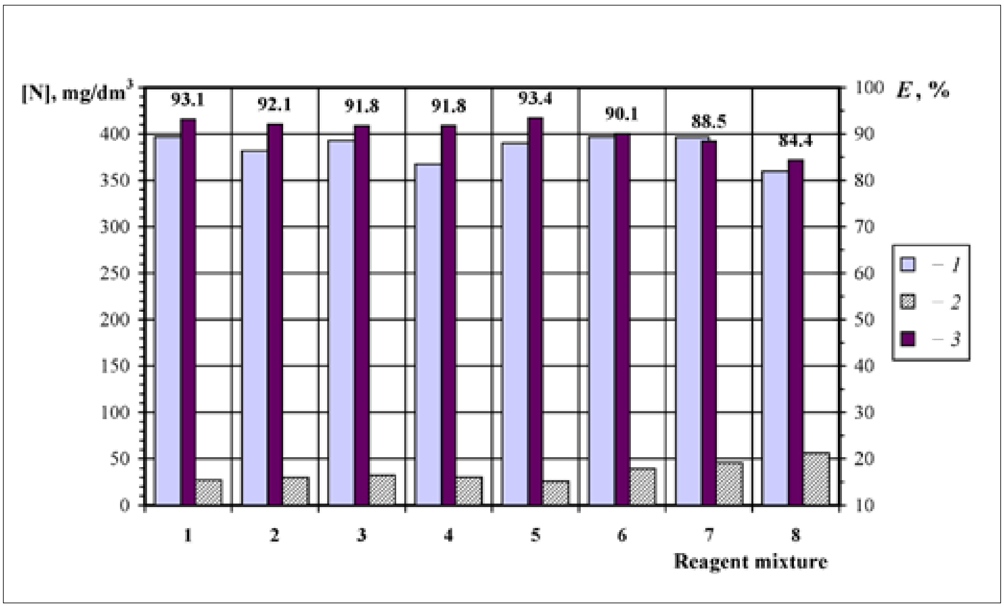

Figure 3. Results of the reagent treatment of Hrybovychi MSW leachate at the pilot-scale treatment plant by the total Kjeldahl nitrogen: 1 – before the reagent treatment; 2 – after the reagent treatment; 3 – treatment effect, %.

reduction significantly exceed corresponding effects for the simple Fenton process, namely 61% obtained by (Deng, 2017), 63% (Bae et al., 1997) and 77–83% (Badawy et al., 2013). It should be noted that the optimum ratio of [H_2O_2]/COD was found to be 0.23–0.25, which is much less comparing the [H_2O_2]/COD=4.4, obtained by (Badawy et al., 2013), and [H_2O_2]/COD=6.8 (Deng, 2007). The significantly higher effects of leachate treatment by COD in the implemented method can be explained by a significant increase in coagulation binding of recalcitrant organic contaminants due to previous flocculation-coagulation treatment using solutions of PAA and aluminium sulphate.

The BOD_5 and BOD_{tot} of Hrybovychi MSW leachate before and after the reagent treatment are presented in Figure 5–6. For the leachate, studied in the pilot-scale treatment plant, the BOD_{tot} value after the aerobic treatment stage was

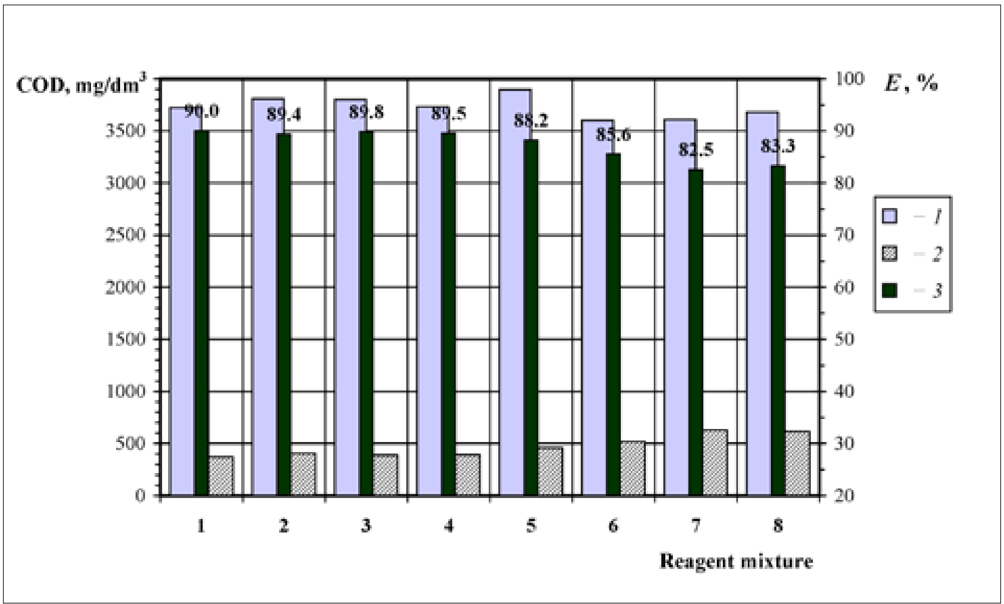

Figure 4. Results of the reagent treatment of Hrybovychi MSW leachate at the pilot-scale treatment plant by COD: 1 – before the reagent treatment; 2 – after the reagent treatment; 3 – treatment effect, %.

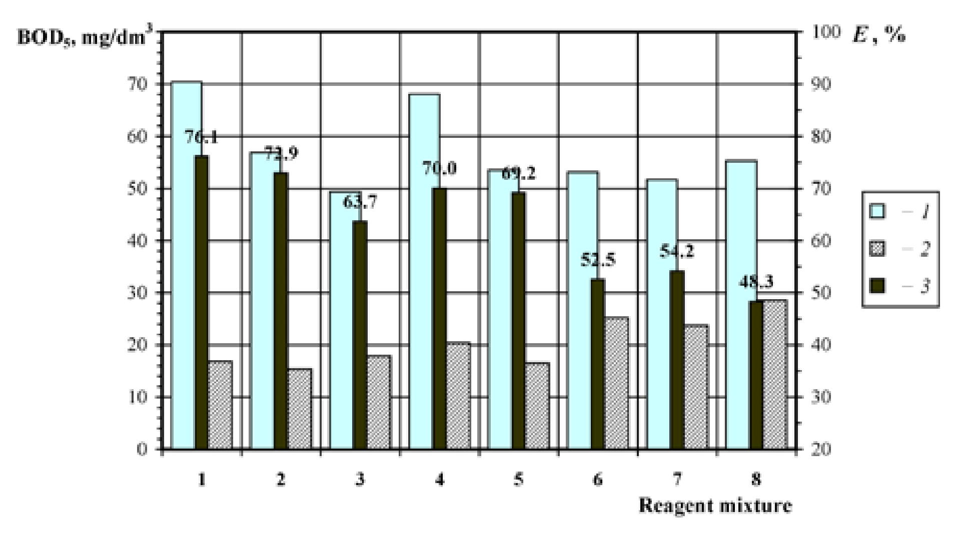

Figure 5. Results of the reagent treatment of Hrybovychi MSW leachate at the pilot-scale treatment plant by BOD_5: 1 – before the reagent treatment; 2 – after the reagent treatment; 3 – treatment effect, %.

consistently much lower than the corresponding value of the limit value of 350 mg/dm^3. BOD_5/COD ratio for treated leachate in cases No.4–No.5 was found to be about 0.04–0.05, similar to less than 0.15 obtained by (Bae et al., 1997), but it is not problem in proposed technology, because reagent treatment follows aerobic biological stage and not vice versa.

Treatment effects by suspended solids obtained in the pilot-scale treatment plant are formally minor and do not exceeded 65% (Fig. 7). However, it should be noted that during the reagent treatment, big quantities of sludge were formed due to the input of coagulant, flocculant and Fenton reagent, so that the actual effects of treatment by suspended solids are much more significant if compare to its total content immediately after the addition of reagent solutions. For none of the reagent mixtures, the suspended solids content in the treated leachate exceeded 120 mg/dm^3, which is much less comparing the respective sewerage discharge limit of 300 mg/dm^3.

An additional difficulty in carrying out the leachate reagent treatment is that high dosages of reagent solutions

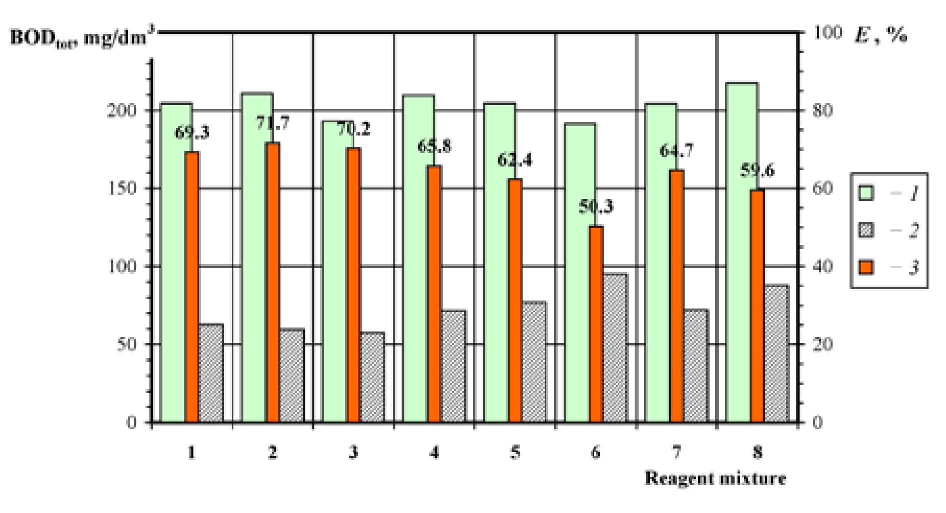

Figure 6. Results of the reagent treatment of Hrybovychi MSW leachate at the pilot-scale treatment plant by BOD_{tot}: 1 – before the reagent treatment; 2 – after the reagent treatment; 3 – treatment effect, %.

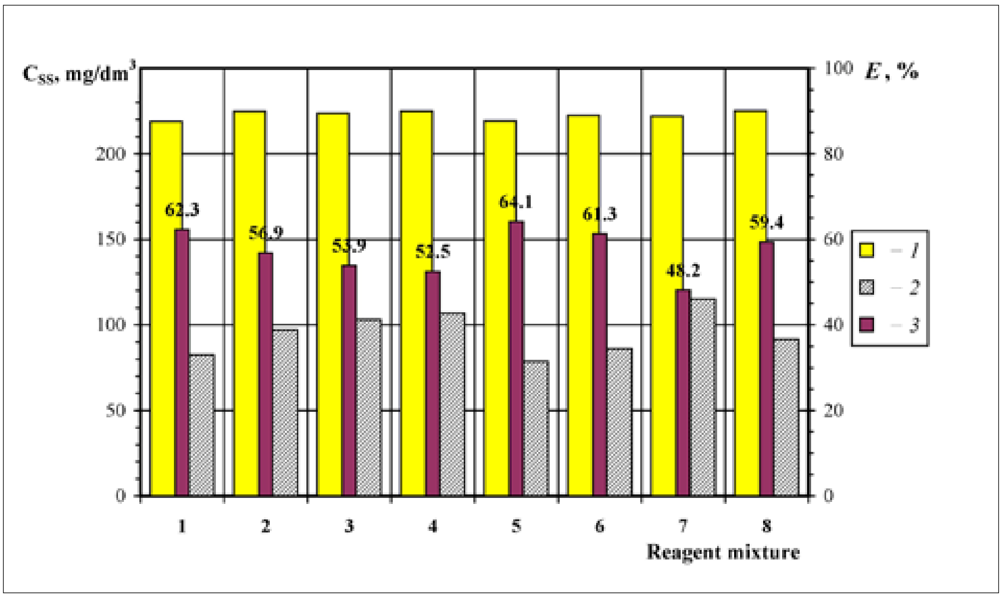

Figure 7. Results of the reagent treatment of Hrybovychi MSW leachate at the pilot-scale treatment plant by suspended solids: 1 – before the reagent treatment; 2 – after the reagent treatment; 3 – treatment effect, %.

cause the acidification of the medium, even threatening to go beyond the allowable range of pH discharge limit (from 6.5 to 9.0). For example, for the reagent mixture No.1, which is characterized by the highest doses of reagents and, in general, the best indicators of other indicators of pollution (e.g., for COD, BOD_{tot} and nitrogen), low pH value 6.26 was obtained, which does not comply with the limits for discharge into the sewerage systems in Ukraine.

The analysis of the results of the pilot-scale study of the leachate reagent treatment suggests that there is a tendency of increasing the content of ammonium nitrogen, TKN, BOD_5, BOD_{tot} and COD in the treated leachate if reagent dosages are decreasing, limiting the minimum allowable dosages of working solutions at the level of reagent mixtures No.4 or No.5. Thus, the optimum dosages of reagent solutions that provide an efficient treatment of Hrybovychi MSW landfill leachate to the Ukrainian limits for discharge into the sewerage system are obtained, namely PAA 0.1 wt% solution – 0.04 m^3 per 1 m^3 of leachate; $Al_2(SO_4)_3$×18 H_2O (10 wt.%) – 0.03–0.04 m^3/m^3; $FeSO_4$×$7H_2O$ (10 wt.%) – 0.06–0.08 m^3/m^3; hydrogen peroxide (10 wt.%) – 0.04–0.05 m^3/m^3.

In general, proposed method of reagent leachate treatment, which is a modified Fenton method, enhanced by preliminary addition of flocculant (PAA) and aluminium coagulant, tested on the pilot treatment plant, meets the requirements for discharge into urban sewerage systems and can be widely implemented at landfill leachate local treatment plants. In the further studies in this direction, it should be clarified how the aeration of the obtained reagent mixtures affects the treatment effects, as well as the possibility of a corresponding reduction in the mass concentrations of flocculant and coagulants in the leachate-reagents mixtures.

4. Conclusions

Landfill leachate reagent treatment using the modified Fenton method, enhanced by preliminary addition of flocculant (PAA) and aluminium sulphate coagulant, was tested on the pilot treatment plant, installed at the Hrybovychi MSW landfill (Lviv region, Ukraine). Reagent treatment of leachate was a second stage of treatment after the aerobic biological pre-treatment. Reagent treatment unit worked in a batch mode, with nominal volume of treated leachate 100 dm^3 per cycle.

Optimal technological parameters of the leachate oxidation using the modified Fenton process, accompanied by the simultaneous coagulation-flocculation and followed by gravitational sedimentation are obtained. The optimum specific dosages of reagent solutions that provide an efficient leachate treatment by key pollutant indicators should be recommended to be equal: PAA 0.1 wt% solution – 0.04 m^3 per 1 m^3 of leachate; $Al_2(SO_4)_3$×18 H_2O (10 wt.%) – 0.03–0.04 m^3/m^3; $FeSO_4$×$7H_2O$ (10 wt.%) – 0.06–0.08 m^3/m^3; hydrogen peroxide (10 wt.%) – 0.04–0.05 m^3/m^3.

High efficiency of COD reduction (88.2–89.5%) is obtained at optimal doses of reagent solutions, namely at a molar concentration 24–30 mmol/dm^3 of hydrogen peroxide and 22–29 mmol/dm^3 of iron (II) ions. The optimum ratio of $[H_2O_2]$/COD was found to be 0.23–0.25.

Obtained maximum effects of COD reduction significantly exceed corresponding effects for the simple Fenton process reported before. This result could be explained by the synergistic effect of additional flocculation and coagulation immediately before the input of Fenton reagent.

The results of the study of the reagent treatment of aerobically pre-treated Hrybovychi MSW landfill leachate at the pilot-scale treatment plant showed the efficiency of the proposed technology and allow recommending this technology for industrial implementation.

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