

Kinetics of the organic matter biodegradation in the leachate from a municipal waste landfill

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ABSTRACT

The paper presents results of the research upon leachate from a municipal waste landfill in north-eastern Poland, where a reverse osmosis process with concentrate spraying on the dumped waste to treat leachate was used. The tests include measurements of oxygen consumption for the mineralization of organic matter in the biochemical oxygen demand (BOD). Measurements were carried out in a 25 d period under laboratory conditions at 20°C and 10°C. The test results were used to estimate the rate constant of biochemical decomposition of organic matter in the leachate. To describe the rate of BOD changes, the assumption was made that the decomposition reaction is the first-order reaction. This assumption was correct when dividing the process into two stages: 0–5 d and 5–25 d. On the basis of the test results, the reaction rate constants for biochemical changes for the oxygen demand in both stages were determined separately. In the first step, reaction rate constants k varied between 0.610 and 0.624 d⁻¹ at 20°C and 0.387 to 0.877 d⁻¹ at 10°C. In the second stage, they were 0.021–0.090 d⁻¹ and 0.009–0.055 d⁻¹, respectively. The R^2 determination coefficient for the calculated values ranged from 0.9903 to 0.9960.

Keywords: Leachate; Reverse osmosis; Concentrate recirculation; Biochemical oxygen demand; Biochemical reaction rate

1. Introduction

Despite the strain to manage waste in a closed cycle, which aims to minimize the amount of waste generated and use of inevitable waste in recycling, in Poland, as in other countries, the main method of disposal of some municipal waste is landfilling. In 2017, 126 million tons of waste were generated in Poland, of which 9.5% was a municipal waste (12 million tons). Of the collected municipal waste, 6.8 million tons were destined for recovery (about 57% of generated waste), and a total of 5.2 million tons were directed to disposal processes, of which 5 million tons (42% of municipal waste generated) were designated for a landfill [1].

One of the problems arising during the operation of landfills is the formation of leachate. Highly variable

composition of leachate depends on several factors, among others: characteristics of the solid waste dumped, degree of separation of the moisture fraction in the municipalities served by the landfill site, seasonality, degree of solid waste stabilization [2–4]. The composition of leachate also depends on technical operations carried out in landfills [5]. Industrial and municipal waste landfill sites create a hazard of groundwater pollution [6].

Due to the unfavorable composition of leachates, their treatment is not easy and creates a problem in choosing the right technological process. Although research on leachate neutralization has been carried out for many years, the development of an efficient and economically justified method of purification remains a current problem in

environmental engineering. The leachate can be treated together with municipal sewage [7,8] or their neutralization is subject to separate, suitably selected technological processes.

For the treatment of leachates characterized by a high biochemical oxygen demand (BOD)/chemical oxygen demand (COD) ratio (>0.5), biological methods are used [9]. Stabilized leachate with a low BOD/COD ratio is most often treated by physical and chemical methods. These methods, in addition to partial removal of contaminants, allow increasing the proportion of biodegradable fraction of organic matter, which enables their more effective purification by biological processes [10–13].

Due to high loads of contaminants and complex composition leachate treatment usually requires various process applications that are expensive and complicated. Hybrid treatment methods: stripping, Fenton, sequential batch reactor, coagulation, coagulation–nanofiltration, coagulation–ultrafiltration–nanofiltration/reverse osmosis, adsorption–ultrafiltration–nanofiltration/reverse osmosis, aerobic activated sludge biological pre-oxidation, coagulation/sedimentation, photo-oxidation through a photo-Fenton reaction combining solar and artificial light, coagulation–filtration–ozonation and many others, are the subject of scientific studies [14–18].

One of the membrane methods applied to neutralize leachate generated during waste storage is the reverse osmosis process [19]. Contaminants contained in the leachate are compacted and removed together with the concentrate, which requires further management. In some landfills, this concentrate is recirculated to the dumped waste, causing changes in the composition of the leachate produced [20] and thus affects the efficiency of the processes used to treat them [21].

The aim of the study is to estimate the rate of biochemical decomposition of organic matter in the leachate from the landfill, in which the leachate was treated by reverse osmosis with recirculation of the concentrate to the landfill. To describe the rate of BOD changes, the assumption was made that the decomposition reaction is the first-order reaction. This assumption was correct after dividing the process into two stages 0–5 d and 5–25 d.

2. Materials and methods

2.1. Landfill characteristic

The landfill site, from which leachate samples were taken, is located in north-eastern Poland. It has been operating since 1983 and covers an area of 25.5 ha. Two cells are intended for landfilling: *A* – with an area of 7.7 ha and *B* – with an area of 5.1 ha. Cell *A* functioned from 1983 to 2010. At that time, about 550,000 tons of waste was collected there. In 2011, the process of its reclamation began. Cell *B* was designed to accept 280,000 tons of waste and has been in operation since 2012. To neutralize the leachate, the operation of the reverse osmosis system was started in 2012. The leachates from cells *A* and *B* in the daily average volume of 50–90 m³, were initially collected in the retention and equalizing reservoir, and then, after acidification with sulfuric acid to pH 6.0–6.5, directed to the sand filter system removing the suspension >50 µm

and cartridge filter with a particle retention capacity >10 µm. After the removal of solid particles, the leachates were fed under pressure to the RO system (ROCHEMCD9-RO disc tube membrane modules with the amount of 1,500 m³ per month). Concentrate from the reverse osmosis process, in 2013 was recirculated to the cell *B*, which was to assure adequate humidity of the deposit and intensification of organic matter decomposition processes. The produced concentrated leachate represents about 25% of the total incoming leachate, that is, up to 375 m³ per month.

2.2. Sample collection and analysis

Studies on the rate of biochemical degradation of organic matter were carried out in three samples taken from the collecting well of leachate – pumping station from cell *B* in the period 06.2016–07.2017. Each sample was averaged from 3 parts collected during the operation of the pumps at intervals of 2 h. Until delivered to the laboratory, the samples were stored at approximately 4°C. The BOD determinations were started immediately after delivering the samples to the laboratory. With the exception of BOD measurement, the determinations were carried out in triplicate, and the results presented are the average of each measurement.

The pH and electrolytic conductivity (EC) were measured *in-situ* using a Hach (USA) HQ40D, a multi-parameter. Other determinations were carried out in laboratory conditions. An Analytik Jena (Germany) TOC multi NC2100 analyzer was used for the total organic carbon (TOC) measurement. The colorimetric methods were used to determine: COD – after 2 h digestion (a K₂Cr₂O₇ method, 620 nm); NH₄-N: salicylate method (655 nm); total phosphorus (TP): molybdovanadate with persulfate acid digestion method (420 nm). Hach DR3900 and DR1900 spectrophotometers were used for colorimetric determinations. Total Kjeldahl nitrogen (TKN) determinations were carried out after mineralization of the sample with sulfuric acid in the presence of a catalyst (K₂SO₄, TiO₂ and CuSO₄). The mineralized sample was steam distilled to the boric acid solution and then titrated with sulfuric acid by the potentiometric method to a pH 4.65. The BÜCHI Labortechnik AG (Switzerland) Kjeldigester K-446, the KjeldMaster K-355 distillation unit and the SI ANALYTICS GmbH (Germany) TitroLine EASY titrator were used for the determinations.

BOD determinations were carried out using the manometric method. The OxiTop, WTW system with the OC110 controller for non-standard BOD measurements, was used. Oxygen consumption measurements were made at two temperatures 10°C and 20°C. For the tests, 50–100 ml of leachate were applied. In order to eliminate oxygen consumption for the nitrification process, a nitrification inhibitor: TCMP-2(chloro-6-(trichloromethyl)pyridine (N-Serve) solution) was added to the samples in an amount sufficient to achieve a concentration of 1.5 mg L⁻¹. Oxygen consumption measurements were recorded at 100 min intervals, for 25 d.

Knowledge of the rate of the biochemical decomposition of organic matter in the leachate can be used during the design of biological treatment technology. In the case of domestic and municipal sewage, calculations of

technological parameters are performed for temperatures of 10°C and 20°C, hence the assumed incubation temperature of the samples.

2.3. Calculations

The rate of organic matter biochemical decomposition was estimated assuming that the mineralization reaction proceeds according to the differential equation of first-order physicochemical reaction:

$$-\frac{dS}{dt} = k \text{ BOD} \tag{1}$$

where BOD is the content of organic substance undergoing biochemical decomposition, expressed as biochemical oxygen demand, mg O₂ L⁻¹, *t* is the reaction time, *d*, *k* is the reaction rate constant, d⁻¹.

After integrating Eq. (1):

$$\frac{\text{BOD}_t}{\text{BOD}_0} = e^{-kt} \tag{2}$$

where BOD₀ is the content of organic substance undergoing biochemical decomposition at the initial incubation time, total BOD, mg O₂ L⁻¹, BOD_{*t*} is the content of organic substance undergoing biochemical decomposition over time *t*, mg O₂ L⁻¹.

Assuming the above, the process of oxygen consumption for the biochemical decomposition of organic matter (*X*) can be written with Eq. (3):

$$X_t = \text{BOD}_0 (1 - e^{-kt}) \tag{3}$$

where *X_t* is the oxygen consumption during the biochemical decomposition of organic matter over time *t*, mg O₂ L⁻¹.

Calculations of the reaction rate constant *k* were made using the least-squares method of errors, after logarithmization of the dependence (2):

$$-k = \frac{\overline{t \cdot \ln(\text{BOD}_i)} - \bar{t} \cdot \overline{\ln(\text{BOD}_i)}}{\overline{t^2} - (\bar{t})^2} \tag{4}$$

$$\ln(\text{BOD}_0) = \overline{t \cdot \ln(\text{BOD}_i)} + k \bar{t} \tag{5}$$

$$\text{BOD}_i = \text{BOD}_0 - X_i \tag{6}$$

where *X_i* is the values of oxygen consumption for the biochemical decomposition of organic matter read during the incubation of samples (BOD measurement reading).

Due to the unknown BOD₀ value, the calculations were made using the subsequent approximation method, assuming subsequent BOD₀ values greater than the maximum BOD read after 25 d, until the ratio of BOD₀ to assumed BOD₀ calculated from Eq. (2) equals 1.00. The calculations were made using the Solver tool in a spreadsheet.

3. Results and discussion

3.1. Leachate characteristics

Basic indicators of leachate contaminants from 2016–2017 are presented in Table 1.

The biodegradability, expressed as BOD₅/COD ratio, is one of the basic parameters characterizing landfill leachates. Leachates from young landfills, in the acidogenic phase, is characterized by a high content of biologically degradable organic matter, and consequently high BOD₅/COD ratio (0.3–0.7). The ratio of BOD₅/COD decreases rapidly with a time of landfill exploitation. Old landfill leachate is characterized by its low ratio of BOD₅/COD (<0,1) [9]. The biodegradability of the leachate taken for analysis ranged from 0.07 to 0.21, which is characteristic of matured landfills.

Parameters of the leachate tested in 2016–2017 differ from those from the same landfill taken in 2013, at the time when recirculation of the concentrate into the bulk waste was carried out [19]. Table 2 presents selected parameters of leachate tested in 2013.

Table 1
Basic parameters of collected leachate

	pH	EC	TOC	BOD ₅	COD	NH ₄ -N	TKN	TP
		mS cm ⁻¹	mg L ⁻¹	mg O ₂ L ⁻¹	mg O ₂ L ⁻¹	mg N L ⁻¹	mg N L ⁻¹	mg P L ⁻¹
Sample 1	8.06	17.25	1,050	739	3,430	1,080	1,210	16.1
	7.91							
	8.03							
Sample 2	7.68	15.45	1,210	347	3,585	921	935	17.2
	7.62							
	7.44							
Sample 3	7.36	15.48	1,250	398	5,364	478	774	16.4
	7.47							
	7.37							
Mean	–	16.06	1,170	495	4,126	826	973	16.6

Table 2
Selected parameters of leachate at 2013 [19]

	pH	EC	BOD ₅	COD	NH ₄ -N	NO _x -N	TN
		mS cm ⁻¹	mg O ₂ L ⁻¹	mg O ₂ L ⁻¹	mg L ⁻¹	mg L ⁻¹	mg L ⁻¹
Min.	7.21	7.68	150	285	94.0	0.01	260
Max.	7.90	9.43	1,350	2,250	899.5	1.55	1,450
Mean	–	8.30	582	1,266	334.7	0.07	567
Median	–	8.24	470	1,286	201.0	0.05	415

TN – total nitrogen

For comparison, leachate quality from municipal landfills at a similar age without concentrate recirculation is given in Table 3.

Recirculation of concentrate into waste results primarily in the accumulation of pollutants in the leachate. In addition, it may cause a change in the rate of decomposition processes taking place in a waste body and a change in the leaching intensity, and as a consequence, a further change in the quality of leachate. The concentrate recirculation may cause a decrease in the leachate pH due to the sulfuric acid addition in the RO technological process [21]. Ions from sulfuric acid and mineral compounds coming from RO concentrate cause a significant increase of EC in the leachate. These changes can be observed immediately after the start of recirculation, but the effect of increasing the EC value is observed in subsequent years, even after completion of recirculation. Increased values of COD and NH₄-N and TKN were also observed in the leachate after 3–4 y from the end of concentrate recirculation to the waste body.

Results of measurements of oxygen consumption for the biochemical decomposition of organic matter in samples 1, 2 and 3 are presented in Figs. 1–6. Figures also show the results of X_t calculations (from equation 3) based on calculated reaction rate constants and BOD₀ values. In addition, for the measurement data, a trend line was applied along with a matching logarithmic function. The R^2 coefficient of determination was given for the values and trend line calculated from the model.

The assumption that biochemical mineralization of an organic substance proceeds according to the differential equation of first order physicochemical reactions was statistically significant when dividing the process into two stages. The first involves a quick decomposition in the first few days (a fixed time of 5 d was assumed). In the second stage, the process of mineralization is slower, among others, due to the lower concentration of the remaining

organic matter decomposable by the biochemical route. Table 4 presents the results of the calculations of the model parameters [Eqs. (2) and (3)]: BOD₀, reaction rate constant k , and determination coefficient R^2 values.

Reaction rate constants k determined at 20°C in the first 5 d had similar values in all analyzed samples and were within 0.610–0.624 d⁻¹. In the second stage, they amounted to 0.021–0.090 d⁻¹. The R^2 determination coefficient calculated for the measured and model values was not lower than 0.99. At the same time, for the empirical values presented in the graphs, the logarithmic trend line $y = a \ln(x) + b$ was determined, with the coefficients a and b and the determination coefficient R^2 . Functions determined in this way were characterized by weaker fitting in relation to the model (2). High fitting of the trend line to the measured values ($R^2 > 0.99$) was obtained only for sample 1.

Studies on the rate of mineralization process carried out by other authors indicate a high variability of reaction rates constants k . According to Fulazzaky [23], in the leachates from various landfills in France, process constants in 5 d fluctuated within 0.0082 (BOD₅ = 485, BOD_{total} = 11,416) to as much as 2.2840 (BOD₅ = 101, BOD_{total} = 198). The obtained R^2 determination coefficients ranged from 0.0016 to 0.9983.

Reaction rate constants k in the first 5 d, estimated on the basis of measurements of oxygen consumption made at 10°C, took values from 0.387 to 0.877 d⁻¹. Between 5 and 25 d, these values were 0.009–0.030. The coefficient of determination calculated for both stages together, as in the calculation for 20°C, was not lower than 0.99. The trend line $y = a \ln(x) + b$ determined for empirical values were characterized by smaller fitting: R^2 was in the range of 0.9144–0.9648.

Although the rate of biochemical decomposition of organic matter affects the leachate treatment process, the research conducted by many authors on changes in the biodegradability of leachate [11,24–28], as well as research on biological leachate treatment [29–31] did not include an

Table 3
Leachate quality from landfills without concentrate recirculation [5,9,22]

	pH	EC	BOD ₅	COD	NH ₄ -N	TN	TP
		mS cm ⁻¹	mg O ₂ L ⁻¹	mg O ₂ L ⁻¹	mg L ⁻¹	mg L ⁻¹	mg L ⁻¹
Min.	6.2	2	600	500	10		5
Max.	6.8	50	2,000	40,000	3,000		100
Mean	–	17	1,000	10,000	800	1,000	30

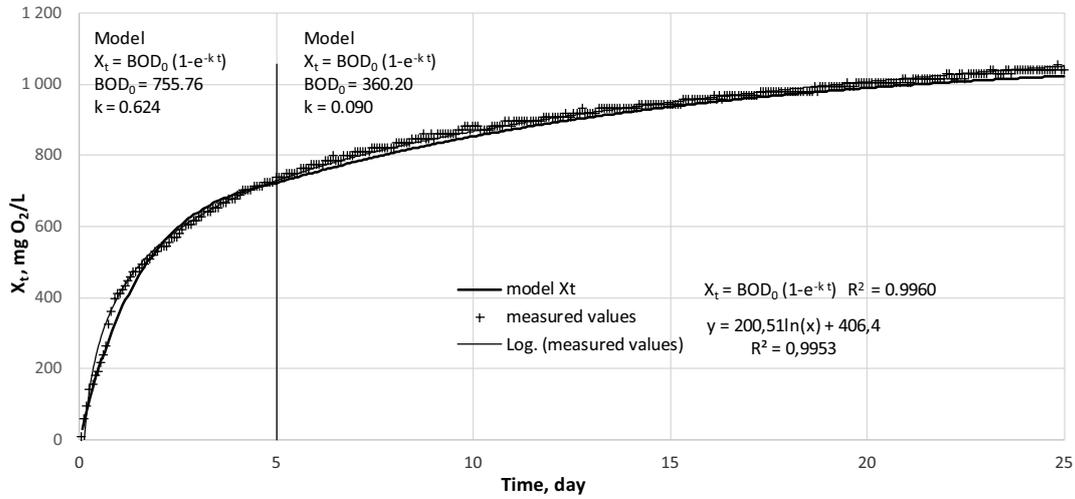


Fig. 1. Biochemical oxygen consumption in leachate sample 1, $T = 20^\circ\text{C}$.

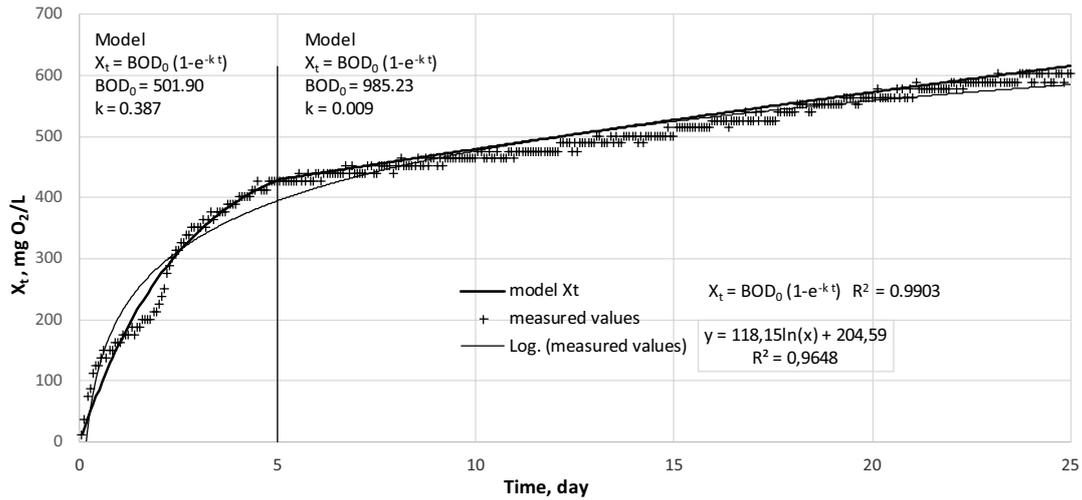


Fig. 2. Biochemical oxygen consumption in leachate sample 1, $T = 10^\circ\text{C}$.

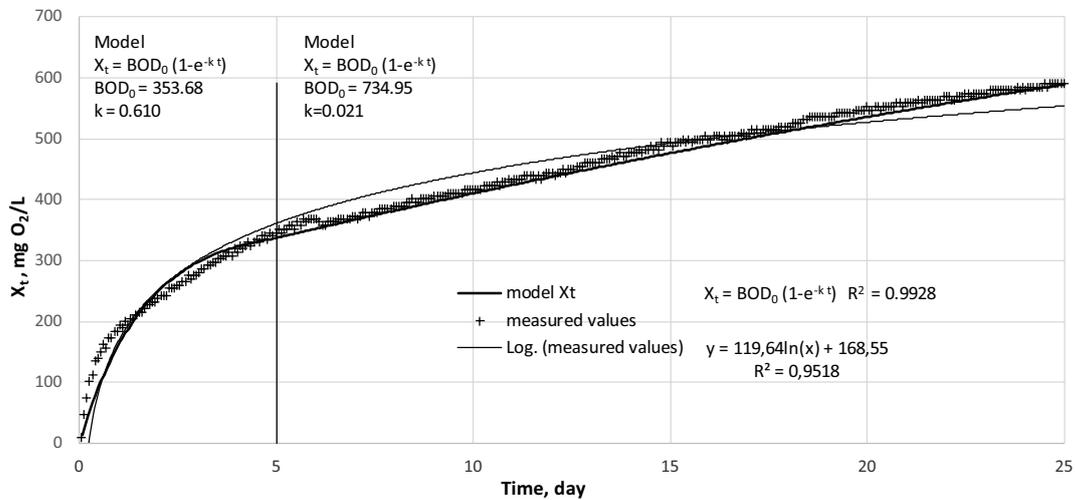


Fig. 3. Biochemical oxygen consumption in leachate sample 2, $T = 20^\circ\text{C}$.

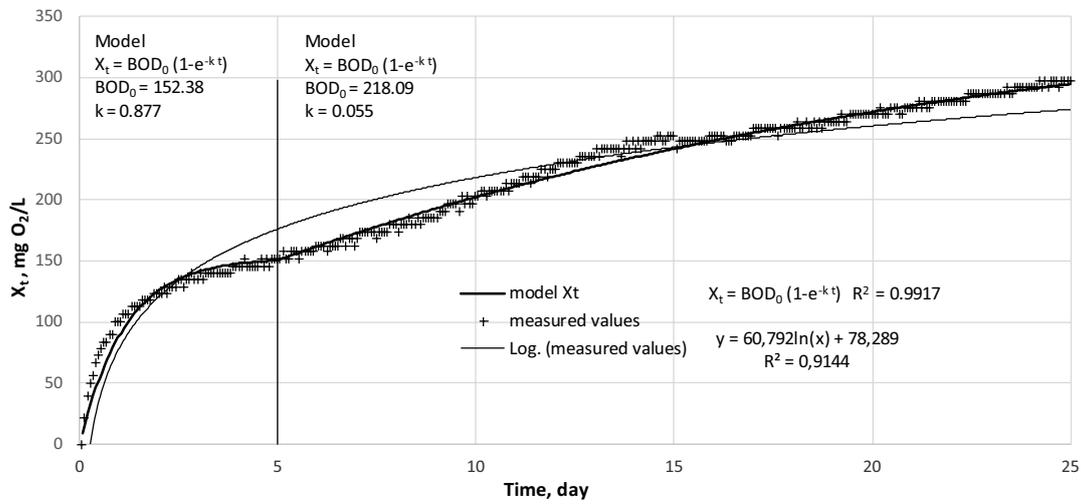


Fig. 4. Biochemical oxygen consumption in leachate sample 2, $T = 10^{\circ}\text{C}$.

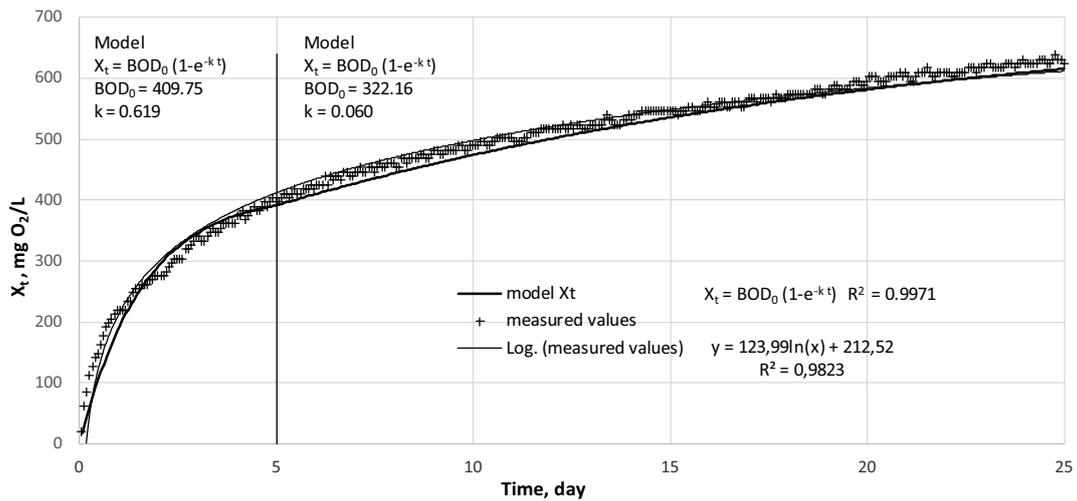


Fig. 5. Biochemical oxygen consumption in leachate sample 3, $T = 20^{\circ}\text{C}$.

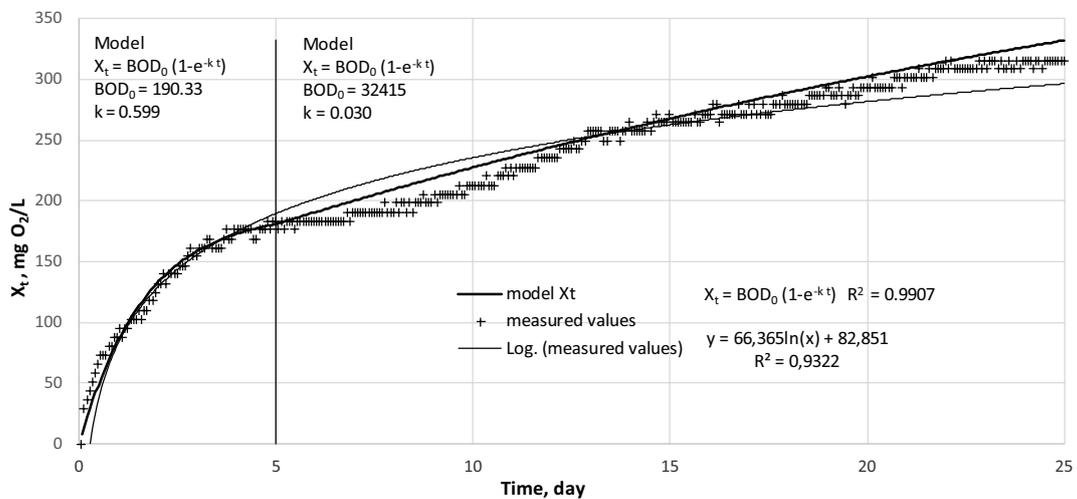


Fig. 6. Biochemical oxygen consumption in leachate sample 3, $T = 10^{\circ}\text{C}$.

Table 4
Values of k , BOD_0 of the different samples

	Sample 1	Sample 2	Sample 3	Sample 1	Sample 2	Sample 3
	20°C			10°C		
Stage I, 0–5 d						
BOD_0 , mg O ₂ L ⁻¹	755.76	353.68	409.75	501.90	152.38	190.33
k , d ⁻¹	0.624	0.610	0.619	0.387	0.877	0.599
Stage II, 5–25 d						
BOD_0 , mg O ₂ L ⁻¹	360.20	734.95	322.16	985.23	218.09	324.15
k , d ⁻¹	0.090	0.021	0.060	0.009	0.055	0.030
Stage I + stage II, 0–25 d						
R^2	0.9960	0.9928	0.9916	0.9903	0.9917	0.9907
BOD_0 , mg O ₂ L ⁻¹	1,116	1,089	731	1,487	370	514
BOD_5 , mg O ₂ L ⁻¹	739	347	398	427	152	184
BOD_5/BOD_0	0.66	0.32	0.54	0.29	0.41	0.36

estimation of the rate of biochemical degradation of organic matter. Knowledge of reaction rate constants may allow a better understanding of the biological leachate treatment process.

4. Conclusions

The tested leachates were characterized by a low degree of biodegradation. The BOD_5/COD ratio was 0.07–0.21. Obtained results of measurements and calculations allow stating that the process of oxygen consumption for the biochemical decomposition of organic matter within 25 d in the tested leachates can be described by the first-order equation for physicochemical reactions after the division of the period into two stages: 0–5 and 5–25 d. In the first step, reaction rate constants k ranged from 0.610 to 0.624 d⁻¹ at 20°C and from 0.387 to 0.877 d⁻¹ at 10°C, while in the second stage, they ranged from 0.021 to 0.090 d⁻¹ and from 0.009 to 0.055 d⁻¹, respectively.

The R^2 determination coefficient for the calculated values ranged from 0.9903 to 0.9960. The model of the mineralization process described by the logarithmic function $y = a \ln(x) + b$ to a smaller extent corresponded to the results of laboratory measurements. The model R^2 fitting ratio was in this case 0.9144 to 0.9953.

The observed oxygen consumption for the biochemical decomposition of organic matter is slightly different at 20°C and 10°C. In the first phase (0–5 d) at higher temperature these changes are practically very similar in each of the tested samples. The k coefficient has similar values, and the R^2 values are the highest. At 10°C, the process differs from the assumed model, although it can still be successfully described mathematically using the assumptions made in the work.

Knowing the basics of microbiology, it is known that the composition of the microorganisms contained in the leachate depends on temperature, and the rate of biological processes depends on this composition. In the case of the analyzed leachate, at a lower temperature, in the first phase of measurements, a smaller amount of organic matter (from 42% to 56%) undergoes biochemical degradation, hence it should be

considered that a higher temperature will be optimal for biological treatment of leachate.

The mathematical description of the rate of oxygen consumption in the second-stage (5–25 d) may be burdened with an error and calculated BOD_0 values (and thus BOD_5/BOD_0) should be treated as an approximation.

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