MIDDLE POMERANIAN SCIENTIFIC SOCIETY OF THE ENVIRONMENT PROTECTION
ŚRODKOWO-POMORSKIE TOWARZYSTWO NAUKOWE OCHRONY ŚRODOWISKA

Annual Set The Environment Protection
Rocznik Ochrona Środowiska

Volume/Tom 20. Year/Rok 2018 ISSN 1506-218X 857-872

Effectiveness of Coagulation in Contaminants Removal from Biologically Treated Landfill Leachate

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1. Introduction

Landfilling is the most used final waste disposal method, because of its lower costs of operation and maintenance. One of the main concerns in landfill site exploitation is production of highly contaminated wastewater – landfill leachates. Highly variable composition of leachate depends on several factors, among others: characteristics of the solid waste dumped, the degree of separation of the moisture fraction in the municipalities served by the landfill site, the seasonality, the degree of solid waste stabilisation and technical treatment at landfills (Kurniawan et al. 2006, Kulikowska & Klimiuk 2008, Szymański & Nowak 2012, Talalaj & Biedka 2015). Municipal and industrial waste landfill sites creates potential hazard of groundwater pollution (Szymański et al. 2017).

To treat landfill leachate several methods are used: leachate transfer (recycling and combined treatment with domestic sewage), biodegradation (aerobic and anaerobic process), chemical and physical methods (chemical oxidation, adsorption, chemical precipitations, coagulation/flocculation, air stripping), membrane process (microfiltration, ultrafiltration, nanofiltration, reverse osmosis) (Renou et al. 2008). One of the method is also landfill leachate treatment in a multi-stage subsurface flow constructed wetland (Wojciechowska 2015, Wojciechowska 2017). The right choice of treatment method depends on the composition of leachate (Fudala-Książek et al. 2016, Nowak et al. 2016).

Complete leachate treatment is complicated, mostly expensive, and generally require various process applications due to high loads of contaminants and complex composition. Hybrid treatment methods: stripping – Fenton – SBR – coagulation (Guo et al. 2010), coagulation – nanofiltration (Mariam & Nghiem 2010), coagulation – UF – NF/RO and adsorption/UF/NF-RO (Dolar et al. 2016), aerobic activated sludge biological pre-oxidation (ASBO) – coagulation/sedimentation – photo-oxidation through a photo-Fenton (PF) reaction combining solar and artificial light (Silva et al. 2017), and many others, are the subject of scientific studies.

Due to its reliability, simplicity and high cost-effectiveness, biological treatment is commonly used for leachate containing high concentrations of BOD. When the BOD/COD ratio has value >0.5, process is very effective in removing organic and nitrogenous matter. Coagulation-flocculation may be used successfully to remove non-biodegradable compounds (Renou et al. 2008).

The aim of this study was to evaluate coagulation-flocculation process in removing contaminants from biologically treated leachate from a municipal landfill.

2. Materials and methods

2.1. Leachate characteristics

The effectiveness of the coagulation process was determined in leachates from the Siedliska landfill near Ełk (north-eastern Poland).The landfill has been in operation since 1983 storing municipal, non-hazardous waste. The facility consists of two cells, of which one (A) is closed and reclaimed, and the other (B) is in operation. In this study leachate from the cell B was used. The cell B has an area of 5.1 ha (enabling the deposition of 280,000 Mg of waste), is operated since 2012 and equipped with drainage. The amount of generated leachate is in the range of 50-90 m^3/d. The leachate is treated in a reverse osmosis system. The RO concentrate is recirculated to the landfill cells, which was supposed to ensure adequate humidity of the bed and intensification of the distribution of organic matter.

The leachate samples for this study were taken in March 2017, directly from the collective drainage well. In laboratory conditions, the leachate underwent a biological pretreatment process in the SBR system. The characterization of raw and biologically treated leachates is presented in Table 1.

Table 1. Properties of raw and biologically pretreated leachates
Tabela 1. Właściwości odcieków surowych i podczyszczonych biologicznie

accuracy	TSS mg L^{-1} (± 1%)	EC mS cm^{-1} (± 0,5%)	TOC mg L^{-1} (± 1%)	BOD_5 mg O_2 L^{-1} (± 1%)	COD_{Cr} mg O_2 L^{-1} (± 1,6%)
Raw	23	15.45	1210	347	3585
Biologically pretreated	84	13.61	1170	50	3390

accuracy	pH (±0,02 pH)	Turbidity NTU (± 2%)	Colour PtCo (± 2%)	TP mg P L^{-1} (± 1,2%)	$P\text{-}PO_4^{3-}$ mg P L^{-1} (± 0,5%)
Raw	7.58	5.1	11842	17.2	9.83
Biologically pretreated	8.94	10.2	10750	16.0	8.71

accuracy	TKN mg N L^{-1} (± 1%)	$N\text{-}NO_3^-$ mg N L^{-1} (± 0,5%)	$N\text{-}NO_2^-$ mg N L^{-1} (± 0,5%)	$N\text{-}NH_4^+$ mg N L^{-1} (± 0,5%)
Raw	935	0.35	-	921.4
Biologically pretreated	168	0.85	-	16.08

2.2. Biological treatment

Biological treatment of leachate was conducted as a bench-scale test, in a sequential biological reactor system (SBR), with nitrification and denitrification assisted by metering of an external carbon source (methanol). The stand comprised a tank with a total volume of 25 L, peristaltic pumps dosing raw sewage and methanol and discharging treated wastewater and biological sludge, mechanical mixer CAT R50D with adjustable speed, aeration system: electromagnetic piston pump Hailea ACO-208 with a diffuser placed on the bottom of the tank, pH, ORP, dissolved oxygen (Hach HQ30D, HQ40D multiparameter) instruments. The operation of the pumps, the mixing and aeration system was controlled automatically, developed individually for the needs of the model by an electronic system based on the ATMEGA328P microcontroller.

The activated sludge used in the reactor came from municipal wastewater treatment plants. Before the actual experiment, it was gradually adjusted to work in leachate for 4 weeks.

The operation cycle of the SBR reactor has been developed in a way that allows obtaining a high degree of ammonium nitrogen removal (average 98.2%) – Table 2.

Table 2. Operating parameters and schedule of SBR reactor
Tabela 2. Parametry i cykl pracy reaktora SBR

Phase	Time / volume
filling	2.0 L
mixing (denitrification)	120 min
mixing and aeration – nitrification	240 min
dozing of metanol	3.0 mL
mixing (denitrification)	180 min
mixing and aeration – nitrification	240 min
dozing of metanol	3.0 mL
mixing (denitrification)	180 min
mixing and aeration – nitrification	240 min
sedimentation	30 min
decantation	1.8 L
removal of excess sludge	0.2 L
duraton of phase, t_C	20.5 h
reaction time, t_R	18.0 h / phase
aeration time, t_{aero}	12 h / phase
active volume, V_R	10.0 L
concentration of active sludge, TS_{BB}	3.5-4.5 mg s.m. / L

The biologically pretreated leachates, as compared to the raw leachates, were characterised mainly by the lower concentration of ammonium nitrogen and Kieldahl nitrogen (the removal rate of 98.3 and 82.1%, respectively). The removal rate of organic matter expressed as BOD, amounted to an average of 85.6%. The remaining indicators in the SBR reactor were removed to a lesser extent (Table 1).

2.3. Coagulation experiment

The coagulation process was carried out by dosing three aluminium coagulants to the leachates: polyaluminium chloride $\{Aln(OH)_mCl_{(3n-m)}\}_x$, aluminium sulphate $(Al_2(SO_4)_3$, (ALS), sodium aluminate ($NaAlO_2$) and one iron coagulant: iron (III) chloride ($FeCl_3$, PIX111). PAX18, ALS, PIX111 were used as ready coagulants produced by Kemipol Ltd., Poland, $NaAlO_2$ – as solution of sodium aluminate. The dose volume of preparations was chosen based on metal ions (mg Me L^{-1}). Each coagulant was added in varying doses – Table 3.

Table 3. Doses of coagulants used in leachate pretreatment process
Tabela 3. Dawki koagulantów stosowane w procesie podczyszczania odcieków

Coagulant	Doses, mg Me L^{-1}				
polyaluminium chloride	125	250	500	750	1000
aluminium sulphate	100	200	400	600	800
sodium aluminate	120	240	480	720	960
iron (III) chloride	200	400	600	800	1000

Before adding the coagulant, the pH of the leachate was adjusted by adding sulphuric acid in an amount sufficient to achieve a coagulation pH in the range 6.0-7.0. The obtained reaction in which the process was carried out is presented in Fig. 1.

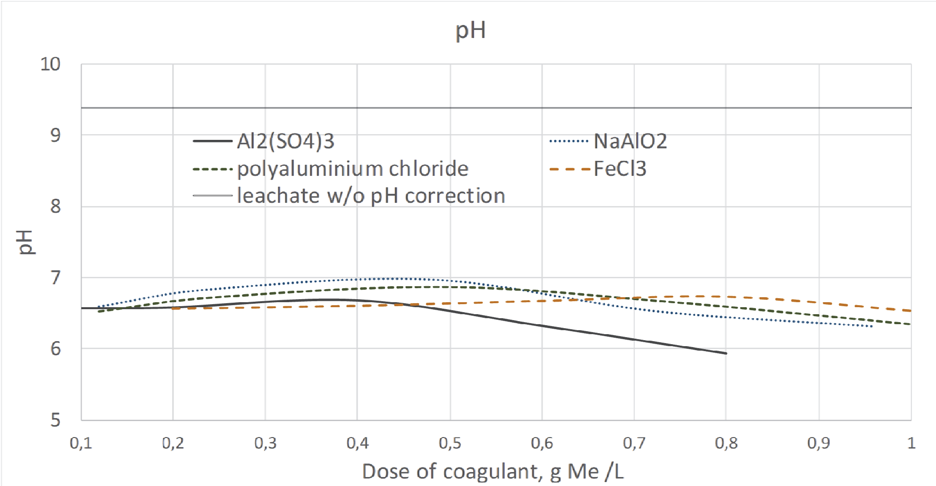

Fig. 1. Changes pH of coagulation process
Rys. 1. Zmiany odczynu pH procesu koagulacji

Coagulation was carried out in cylindrical glass beakers, in 0.5-liter samples. The process included two mixing phases: rapid (150 RPM) for 30 seconds and slow (20 RPM) for 30 min. A Velp Scientifica JLT 6 flocculator was used for mixing. After the mixing was completed, the samples were allowed to settle for 4 hours. Liquid from a height of approx. 2 cm from the surface was taken for testing (sample volume: 100 mL).

2.4. Analytical methods

Several analytical parameters were selected: pH, conductivity (mS/cm), colour (PtCo), turbidity (NTU), chemical oxygen demand (COD, mg O_2 L^{-1}) phosphorus total (TP, mg P L^{-1}). The conductivity and the pH were measured by a conductivity and potentiometric method, respectively, using a portable pH meter (HACH HQ40). The chemical oxygen demand was analyzed using a colorimetric method with a HACH spectrophotometer (620 nm) after a 2-hour reactor digestion (a $K_2Cr_2O_7$ method). The phosphorus total was measured using test'N Tube Vials (molybdate method) on the HACH spectrophotometer (880 nm) after a 1-hour acid persulfate digestion. Colour was analyzed by 455 nm (HACH). For turbidity measurement, the WTW Turb® 550 turbidity meter was used. The obtained results were the mean value of three determinations carried out simultaneously.

3. Results and discussion

The conducted research allowed to determine the degree of removal of selected indicators of leachate contamination depending on the coagulant used and its dose. The reaction of the process was to be maintained in the range of 6.0-7.0, which is higher than optimal for removing COD and turbidity. This assumption resulted from the potential for further leachate purification in membrane processes, in which the capacity of the membranes is reduced by a high sulphate content (Wiszniowski et al. 2006). The actual pH values are presented in Fig. 1. In the assessment of the effectiveness of the coagulation process, selected indicators were taken into account, i.e. the concentration of organic matter, colour, turbidity and total phosphorus.

Organic matter

Due to the concentration of easily and non-easily decomposable organic matter expressed as a BOD/COD ratio, which amounting to 0.098 in raw leachate, these leachates should be classified as stabilized (Kang et al. 2002). In biologically pretreated leachates, the value of this ratio was 0.015. Due to the low BOD values, only the COD indicator was taken into account in the assessment of the coagulation process.

The degree of removal of organic matter, using the above-mentioned doses of metal ions, reached approx. 60% in the case of aluminium coagulants, and less than 40% in the case of iron coagulant. However, a different molar concentration of iron and aluminium with the same weight concentration of metals should be taken into account, which for aluminium is 2.07 times higher. The dependence of the indicator removal rate on the coagulant dose, for aluminium sulphate, polyaluminium chloride and ferric chloride, was similar to linear. The dose of sodium aluminate up to approx. 0.5 g Al L^{-1} allowed the COD to be reduced by about 20%, only higher doses allowed for more effective removal of organic matter.

In the work of other authors (Silva et al. 2004, Rivas et al. 2003, Monje Ramirez & Orta de Velasquez 2004, Tatsi et al. 2003, Leszczyński 2011), the effectiveness of COD removal in the coagulation process was estimated in the range of 10-90%. At the dose of iron and aluminium coagulants amounting to approx. 18 mmol Me L^{-1}, there were no clear differences between them in the effectiveness of COD removal. When treating old landfill leachate applying coagulation –flocculation with iron trichloride or with aluminium polychloride, it was possible to reduce the non-biodegradable organic matter by 73-62%, also reducing turbidity and colour by more than 97% (Castrillón et al. 2010). In the combined treatment system including a coagulation process, followed by a Fenton oxidation, obtained result of COD removal was 65.3% (Barbusiński & Pieczykolan 2010).

Colour

The use of polyaluminium chloride and aluminium sulphate gave a similar effect in colour removal. The dependence of the degree of colour removal on the applied dose of coagulant (similarly as in the case of COD) was similar to the linear one. At the highest dose, the values of 89.8 and 89.9% were achieved, respectively. The use of sodium aluminate at a dose of up to 0.5 g Al L^{-1} resulted in a slight degree of colour removal (by approx. 20%), only a higher dose (0.96 g Al L^{-1}) enabled colour removal in 89%. Iron (III) chloride at a dose of 0.2 g Fe L^{-1} caused a colour increase, only doses above 0.6 g Fe L^{-1} allowed a reduction of this index (up to 54.8% at a dose of 1.0 g Fe L^{-1}). Similar degrees of colour removal from landfill leachate (55 ÷ 70%) were obtained by Zamora at al. (2000), at doses of $Al_2(SO_4)_3$ + $FeCl_3$ of 0.738 + 1.136 g L^{-1}. Using doses of $FeCl_3{\cdot}6H_2O$ (10 g/L at pH 8.0) and $Fe_2(SO_4)_3\ {\cdot}7H_2O$ (12 g/L at pH 7.5) Liu et al. (2012) obtained 93.31% and 74.65% colour removal, respectively. Investigation the efficiency of coagulation and flocculation processes for removing colour from a semi-aerobic landfill leachate from one of the landfill sites in Malaysia (Aziz et al. 2006) using four types of coagulant namely aluminium (III) sulphate (alum), ferric (III) chloride, ferrous (II) sulphate and ferric (III) sulphate result that ferric chloride was superior to the other coagulants and removed 94% of colour at an optimum dose of 800 mg/l at pH 4. The effect of coagulant dosages on colour removal showed similar trend as for COD, turbidity and suspended solids.

Phosphorus total

The biological treatment process used in the experiment, optimized to remove nitrogen compounds, did not allow achieving a high total phosphorus removal rate of 7%. By coagulation, this contamination is removed from the leachate usually with high efficiency, because of forming by orthophosphate sparingly soluble complexes with coagulants.

In the case of biologically treated leachates, the use of four coagulants has resulted in a different removal effect of phosphorus compounds. A high degree of removal was already observed at the smallest doses of polyaluminium chloride, sodium aluminate and iron chloride. It amounted to 54.7%, 67.2% and 68.7% respectively. Increased doses allowed further phosphorus removal to reach the maximum values of 96.9%, 95.3% and 82.8%. The dependence of the effectiveness of aluminium sulphate in the removal of phosphorus on the applied dose was similar to the linear course, as in the case of other indicators – Fig. 2.

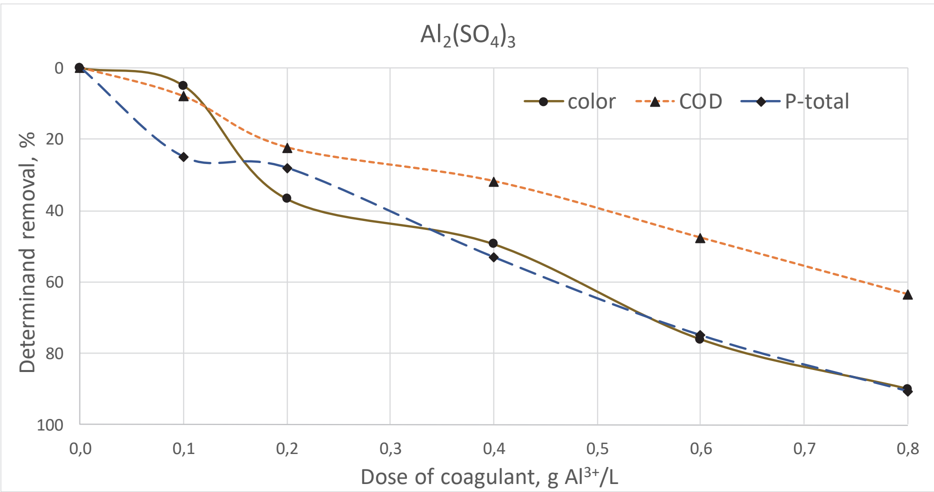

Fig. 2. The effect of contaminants removal depending on the applied dose of aluminium sulphate
Rys. 2. Stopień usunięcia zanieczyszczeń w zależności od zastosowanej dawki siarczanu glinu

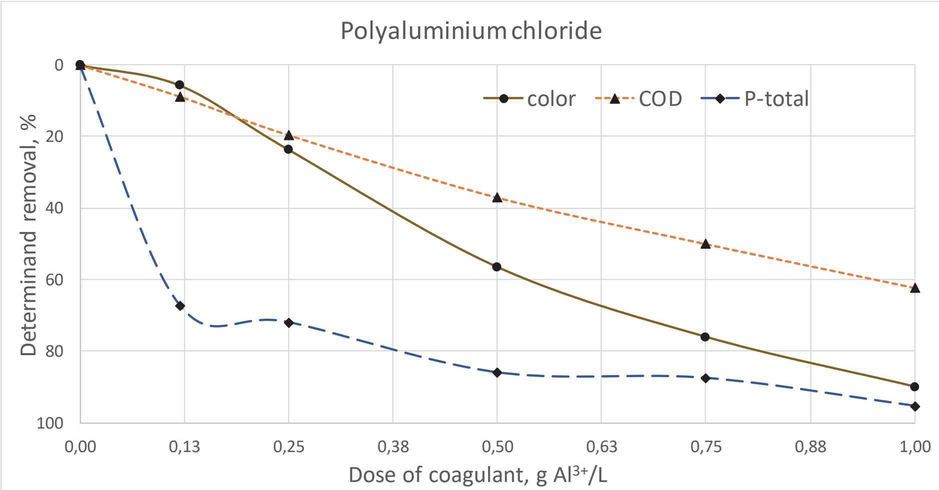

Fig. 3. The effect of contaminants removal depending on the applied dose of polyaluminium chloride

Rys. 3. Stopień usunięcia zanieczyszczeń w zależności od zastosowanej dawki chlorku poliglinu

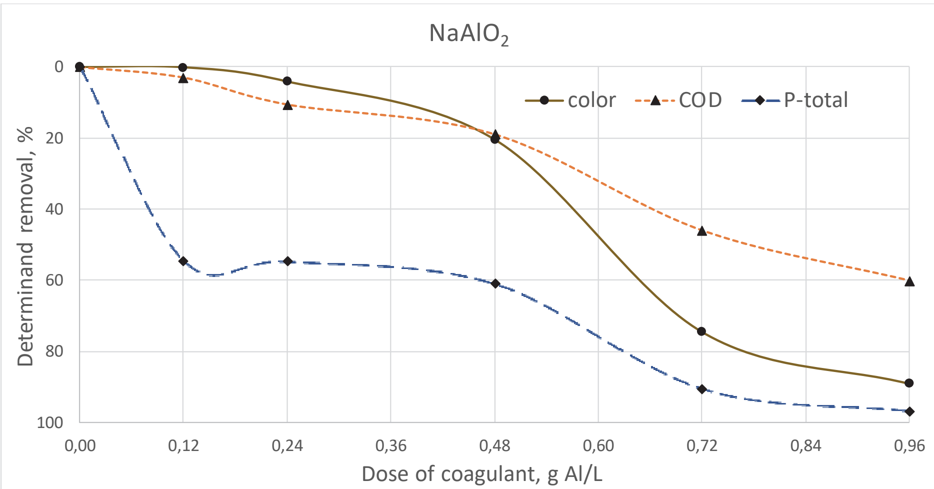

Fig. 4. The effect of contaminants removal depending on the applied dose of sodium aluminate

Rys. 4. Stopień usunięcia zanieczyszczeń w zależności od zastosowanej dawki glinianu sodu

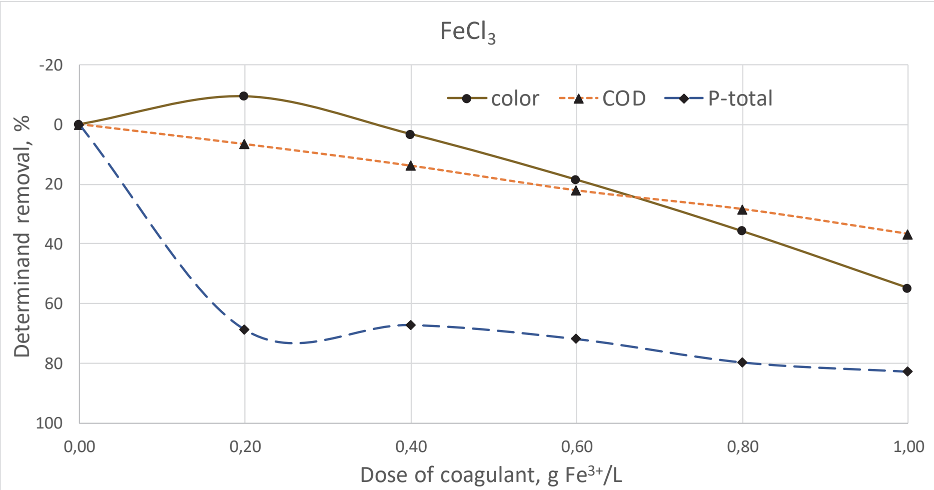

Fig. 5. The effect of contaminants removal depending on the applied dose of iron chloride (III)
Rys. 5. Stopień usunięcia zanieczyszczeń w zależności od zastosowanej dawki chlorku żelaza (III)

Turbidity

The biologically treated leachates manifested twice higher turbidity (10.2 NTU) than the raw leachate (5.1 NTU). These values are smaller than those presented in other works (Tatsi et al. 2003). The addition of aluminium coagulants in doses lower than the maximum adopted in the described experiment caused an increase in turbidity, that is the opposite of those in the case of leachates characterized by higher turbidity (Tatsi et al. 2003). Only higher doses (aluminium sulphate – 0.8 g Al L^{-1}, polyaluminium chloride – 0.75 g Al L^{-1}, sodium aluminate – 0.96 g AL^{-1}) allowed obtaining clear leachates with turbidity lower than initially. The maximum turbidity was observed at doses of: aluminium sulphate – 0.2 g Al L^{-1}, polyaluminium chloride 0.25 g Al L^{-1}, sodium aluminate 0.72 g Al L^{-1}. Additive iron (III) chloride in all applied doses caused an increase in turbidity – Fig. 6. Using doses of $FeCl_3 \cdot 6H_2O$ (10 g/L at pH 8.0) and $Fe_2(SO_4)_3 \cdot 7H_2O$ (12 g/L at pH 7.5) Liu et al. (2012) obtained 98.85% and 94,13% turbidity removal from landfill leachate, respectively. High turbidity removal efficiencies may be obtained in coagulation flocculation (CF) process. Doses of coagulant (ferric chloride) and flocculant (cationic polymer): 4.4 g/L and 9.9 mL/L respectively, reached

69±4.8% removal in leachate treated by anaerobic process (Bakraouy et al. 2017).

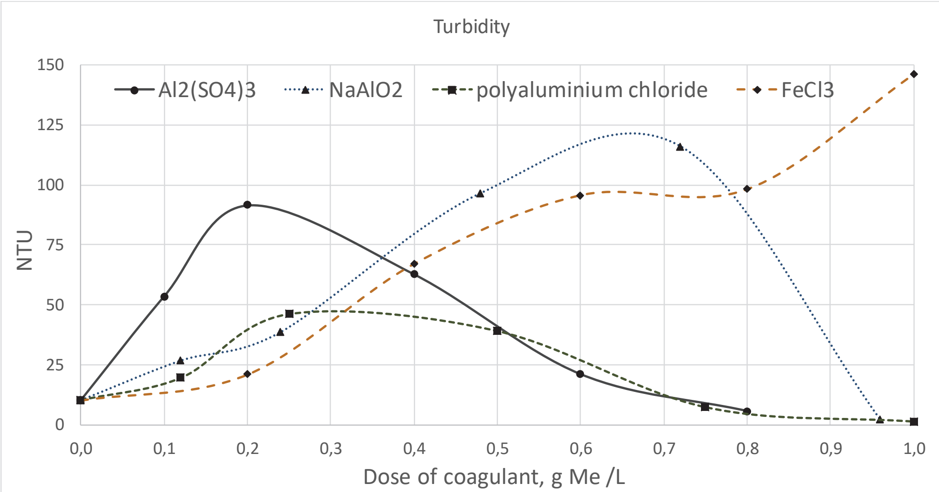

Fig. 6. Changes in turbidity of leachates at different doses of coagulants
Rys. 6. Zmiany mętności odcieków w zależności od dawki koagulantu

4. Conclusions

Treatment of landfill leachates, due to the degree of their contamination, usually requires the use of several technological processes. Obtained results of the tests carried out on biologically pre-treated leachate indicate the possibility of using a coagulation process to partially remove the remaining contamination. Leachates thus prepared, devoid of most nitrogen compounds in the biological process and organic matter that are difficult to decompose in the coagulation process, can then be directed, for example, to membrane processes.

Coagulation of stabilised, biologically pretreated leachates, allows for a high removal rate of organic matter. When aluminium coagulants were used in the doses of 0.8-1,0 g Al L^{-1}, a removal effect of more than 60% was obtained, whereas with iron coagulate (Fe^{3+}) in the dose of 1 g Fe L^{-1}, the effect was lower and amounted to only 36.6%. A high removal degree, reaching 89.0-89.9% using aluminium coagulants and 54.8% in the case of iron coagulant was also obtained for the colour, at the maximum doses used in the experiment.

All the substances used enabled obtaining a high degree of phosphorus removal: 82.8% for iron (III) and 90.6-96.8% for aluminium.

High doses of coagulants resulted in the formation of large amounts of sludge that must be removed before being subjected to subsequent pretreatment processes. Maintaining the assumed pH of the coagulation process carried out using sodium aluminate required the use of increased volumes of H_2SO_4. A similar effect of leachate treatment with the use of other aluminium coagulants, without the need to introduce significant amounts of additional substances, leads to conclusion that the aluminate is less effective in leachate pretreatment.

The paper was prepared during scientific project No. S/WBiIS/2/2014 financed by Polish Ministry of Science and Higher Education.

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Efektywność procesu koagulacji w usuwaniu zanieczyszczeń z odcieków składowiskowych oczyszczonych biologicznie

Streszczenie

W artykule zaprezentowano wyniki badań skuteczności procesu koagulacji w usuwaniu zanieczyszczeń z podczyszczonych biologicznie odcieków pochodzących ze składowiska odpadów komunalnych. Odcieki podczyszczone biologicznie charakteryzowały się niskim stężeniem NH_4^+–N (16,08 mg N L^{-1}), oraz TKN (167,8 mg N L^{-1}). Zawartość substancji organicznej wyrażonej jako BZT_5 wynosiła 50 mg O_2 L^{-1}, stężenie związków organicznych trudno rozkładalnych ChZT wynosiło 3390 mg O_2 L^{-1}. Proces koagulacji prowadzono w warunkach laboratoryjnych, przy zastosowaniu zmiennych dawek koagulantów glinowych: polyaluminium chloride, aluminium sulfate, sodium aluminate oraz żelazowego: iron (III) chloride. Maksymalne dawki wynosiły 0,8-1,0 g Me L^{-1}. pH procesu utrzymywało się w granicach 5,93-6,97. Efekt usunięcia substancji organiczniej (ChZT) uzyskano na poziomie 60-63% dla koagulantów glinowych oraz 37% dla iron (III) chloride. Zmniejszenie barwy nastąpiło o 90%, a zawartość fosforu całkowitego obniżona została maksymalnie o 97%.

Abstract

The effectiveness of the coagulation process in removing contaminants from biologically treated leachates from a municipal waste landfill was investigated. The biologically treated leachates were characterized by a low concentration of NH_4^+-N (16.08 mg N L^{-1}), and TKN (168 mg N L^{-1}). The organic matter concentration, expressed as BOD (Biological Oxygen Demand) was 50 mg O_2 L^{-1}, the concentration of hard-to-break COD (Chemical Oxygen Demand) organic compounds was 3390 mg O_2 L^{-1}. The coagulation process was carried out in laboratory conditions, using variable doses of aluminium coagulants: polyaluminium chloride, aluminium sulphate, sodium aluminate and iron: iron (III) chloride. The maximum doses were 0.8-1.0 g Me L^{-1}. The process was maintained between 5.93-6.97. The removal effect of the organic matter (COD) was obtained at 60-63% for aluminium coagulants and 37% for iron (III) chloride. The colour reduction occurred in 90% and the total phosphorus concentration was reduced to a maximum of 97%.

Słowa kluczowe:
odcieki, oczyszczanie biologiczne, koagulacja

Keywords:
leachate, biological treatment, coagulation