## **CHAPTER IV**

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# TREATMENT TECHNOLOGIES OF MUNICIPAL WASTE LANDFILL LEACHATES

### Introduction

For many years now, one of the most important problems in the area of environment protection consists in the waste management defined as the whole of activities aimed at reduction of quantity, development of effective methods of utilization, and neutralization of waste.

Waste utilization by means of landfilling creates a number of hazards to the environment. Leachates can be counted among them.

The main source of leachates consists in penetration of surface and underground waters as well as external water. Water from a landfill is emitted in the form of vapor and as a component of the landfill gas. Water not absorbed by the waste generates leachates that accumulate at landfill bottom or fill empty spaces at different landfill levels in the form of the so-called suspended water (OBRZUT 1997). The leachates are created when the damp content in the landfill bed exceeds its retention capacity defined as the maximum water quantity that can be retained in porous material of the landfill (EL-FADEL et al. 2002).

Leachate composition reflects microbiological activity of a landfill. Leachates represent a complex and variable mixture of organic, inorganic and microbiological substances and suspensions of solid substances in water. Leachate composition depends also on waste type, e.g. plaster or gypsum can be transformed by anaerobic bacteria into sulfides. However, the most important factor affecting leachate composition is the landfill age (OBRZUT 1997, EL-FADEL ET AL. 2002, OZKAYA 2005).

Municipal waste and some industrial waste, neutralized by means of landfilling, contain mixtures of hazardous substances of various types. Their sources include unwanted products disposed to landfill sites after being used in households or industrial enterprises. As a result of long stay in a landfill bed, they are subject to partial degradation. Consequently, a number of potentially hazardous substances occur in leachates. Many of them demonstrate xenobiotic nature. According to SLACK et al. (2005), more than 200 organic substances were identified up to date in leachates from municipal waste landfill sites, including 35 substances considered potentially hazardous. On the other hand, in ground waters existing in areas surrounding waste landfills, more than 1000 substances of different types were

detected. This means that transformation and/or partial degradation of hazardous substances of various types lead to release of intermediate products and can result in contamination of ground waters.

#### Chemical transformations in leachates in waste dumping period

KURNIAWAN et al. (2006), and earlier KANG et al. (2002), have classified landfill sites, depending on period of their operation, as: young; those in the phase of maturation and stabilization (medium-aged); and stabilized (old).

It follows from numerous published reports that in a landfill's initial operation phase, leachates contain organic substances that are relatively easily subject to biochemical transformations in biological wastewater treatment plants. With increasing landfill age, content of simple organic compounds decreases and highmolecular-weight compounds with little susceptibility to biodegradation first come out, and finally dominate in leachates. According to SURMACZ-GÓRSKA et al. (2000), in case of organic substance content expressed as COD being low and not exceeding 2.000 mg·dm<sup>-3</sup>, one deals with compounds that are hard to decompose biologically. Simultaneously with biochemical transformations. processes of adsorption, dissolution, dilution, ion exchange and precipitation occur as a result of which concentration of organic and inorganic substances varies in time (TREBOUET ET AL. 2001, KANG ET AL. 2002, TATSI ET AL. 2003, RIVAS ET AL. 2004, OZKAYA 2005).

With increasing landfill age, strong decrease of both  $BOD_5$  and COD values can be observed as well as reduction of  $BOD_5$ :COD ratio in leachates, according to data available in the literature (Table 1).

It follows from studies carried out by KACZOREK, LEDAKOWICZ (2002) and SURMACZ-GÓRSKA et al. (2000) that concentration of ammonia nitrogen in leachates from domestic waste landfill sites reaches the value of 3,000 mg·dm<sup>-3</sup>. OBRZUT (1997) reports that ammonia nitrogen concentration in leachate samples taken from ten landfill sites in Poland varied from 1.7 to 1,520 mg·dm<sup>-3</sup>, with the average value of 398 mg·dm<sup>-3</sup>. Studies carried out by KULIKOWSKA (2002) revealed that concentration of ammonia nitrogen increased together with increasing landfill age within the first five years of operation from about 100 mg·dm<sup>-3</sup> up to 600 mg·dm<sup>-3</sup>.

An attempt to determine the nitrogen content in leaches from mature and stabilized landfills was carried out by other authors. EL-FADEL et al. (2002) report that together with increasing landfill age, ammonia nitrogen content in leachates decreases from  $1,500 \text{ mg} \cdot \text{dm}^{-3}$  in the methane fermentation phase to less than 50 mg $\cdot \text{dm}^{-3}$  in stabilization phase. Similar trend was observed by KANG et al. (2002). In the period of first ten years of landfill operation, ammonia nitrogen concentration remained at virtually constant level of  $1,826-1,896 \text{ mg} \cdot \text{dm}^{-3}$ , and then decreased down to the value of  $892 \text{ mg} \cdot \text{dm}^{-3}$ .

Sources of heavy metals in leachates include industrial waste such as ashes, batteries, dyes etc. (ERSES, ONAY 2003) as well as municipal waste containing

electronic parts, fluorescent lamps, thermometers, batteries, pesticides and other (WARD et al. 2005).

Table 1

Landfil l age [years]	Value			
	Parameter			References
	COD	BOD	BOD/COD	References
	[mg <sup>·</sup> dm <sup>-3</sup> ]	[mg <sup>·</sup> dm <sup>-3</sup> ]		
< 5	2640	600	0.22	Lo i in. (1996)
	1727	1058	0.61	SURMACZ-GÓRSKA i in. (2000)
	1183	331	0.28	SURMACZ-GÓRSKA i in (2000)
	41507	32790	0.78	KANG i in. (2002)
	15000-40000	10000-25000	0.6	EL-FADEL i in. (2002)
	480 - 1801	76 - 721		KULIKOWSKA (2002)
	3000-15000	300-15000	0.1-1	KURNIAWAN i in. (2006)
5-10	1660 - 1700	100 - 160	0.06-0.09	Lo i in. (1996)
	2150	215	0.1	TREBOUET i in. (2001)
	10000-20000	1000-4000	0.1-0.2	EL-FADEL i in. (2002)
	5348	2684	0.5	KANG i in. (2002)
	< 3000	< 300	< 0.1	KURNIAWA i in. (2006)
> 10	550	16.5	0.03	TREBOUET i in. (200)]
	1000 - 5000	50 -1000	0.05 - 0.2	EL-FADEL i in. (2002)
	1367	145	0.1	KANG i in. (2002)
	7400 - 8800	475	< 0.1	RIVAS i in. (2004)
	2422 - 3945	106 - 195	0.03 - 0.05	BILA i in. (2005)
> 20	< 1000	< 50	< 0.05	EL-FADEL i in. (2002)

Concentration of organic compounds in leaches versus landfill age measured by means of BOD and COD indicators

Solubility and mobility are closely related to transformations occurring in the landfill and depend on reaction value, redox potential, and, moreover, on presence of organic and inorganic substances able to form complexes (BOZKURT et al. 2000).

The highest concentrations of metals were detected in leachates from young landfills still in the acid fermentation phase, when the reaction value was low. In both maturation and stabilization phases, the reaction value becomes neutral and solubility of metals decreases (ERSES, ONAY 2003).

With increasing landfill age, change in type of organic matter degradation products occurs — from low-molecular-number volatile organic acids to fulvic and humus acids. Solubility of high-molecular-weight acids is different, but most of them demonstrate metal sorption ability. Metals adsorbed on surfaces of humus substances may therefore appear in both colloidal and suspension fractions.

In the landfill's maturation phase, insoluble metal sulfides are created easily. Metals may be also precipitated in the form of carbonates, hydroxides, and even phosphates (ERSES, ONAY 2003).

SLACK et al. (2005) report that only 0.02% of heavy metals dumped on a landfill site penetrate to leachates within the period of 30 years. Similarly, BOZKURT et al.

(2000) claim that more than 99.9% of metals are retained in the landfill bed as a result of sorption on both organic and inorganic molecules (e.g. iron hydroxy oxides) and precipitation. Similar observations were made by other authors (AL-YAQOUT, HAMODA 2003, ERSES, ONAY 2003, WARD et al. 2005). In some cases, concentration of metals can be quite high. SLACK et al. (2005) report that concentration of zinc can reach the level of up to 1000 mg·dm<sup>-3</sup>, that of nickel — 13 mg·dm<sup>-3</sup>, copper — 10 mg·dm<sup>-3</sup>, and lead — 5 mg·dm<sup>-3</sup>. ROBINSON et al. (2005) demonstrated that upper limit of chromium concentration can reach the value of 13.1 mg·dm<sup>-3</sup>. Concentration of mercury in leachates can be even as high as 2 mg·dm<sup>-3</sup> (BILA et al. 2005).

Presence of organic and inorganic hazardous substances and, in many cases, also unidentified products of degradation of organic substances, is the cause of leachate toxicity. In the opinion of CLÉMENT et al. (1997), ammonia nitrogen, heavy metals (Ag, Hg, Pb, Cd, Mn, Zn and Cu) and organic compounds such as tannins, lignin and phenols, can result in toxicity of leachates.

Studies on toxicity of leachates are rather rare. To date, the following groups of organisms were used for this purpose: destroyers — *Vibrio fisheri*; producers — *Scenedesmus subscapitatus*, *Lemna minor*, *Selenastrum capricornutum*; and consumers — *Brachionus calciflorus*, *Daphnia magna*, *Thamnocephalus platyurus* (CLÉMENT et al. 1997, SILVA et al. 2004, MERIÇ et al. 2005).

### **Determinants of the research**

In recent years, intensive research work is carried out on treatment of landfill leachates. For the studies, leachates from municipal waste landfill site in Wysieka near Bartoszyce (Warmińsko-Mazurskie province) were used. The work was carried out in the period when stabilization of biochemical processes in the landfill bed occurs typically. Total nitrogen concentration in leachates was then at the level of 749 mg·dm<sup>-3</sup>, and that of ammonia nitrogen — 636 mg·dm<sup>-3</sup>.

Technological research work was carried out simultaneously on four research stands in reactors denoted as SBR 1, 2, 3 and 4. The leachate retention time was fixed and amounted to 3 d, with the cycle period of 24 h.

Reactors SBR 1 and 3 contained active sludge, while reactors SBR 2 and 4 were filled with active sludge and stationary packing suspended below leachate surface. The packing consisted of 42 strips of PVC sponge with dimensions of  $2 \times 11$  cm. The operational cycle of SBR 1 and 2 reactors included filling, aeration, sedimentation and aeration phases (reactor's aerobic operation conditions), while operation of SBR 3 and 4 reactors consisted of filling, stirring, aeration, sedimentation and decantation phases (anaerobic-aerobic conditions).

In order to determine effectiveness of the process, the following contamination indicators were controlled in the SBR reactors' inlets and outlets: ammonia nitrogen, nitrate nitrogen, and nitrite nitrogen (HERMANOWICZ et al. 1999).

For the purpose of further removal of organic substances from biologically treated leachates, they were subject to further chemical treatment with the use of Fenton's reagent. The research work on leachate treatment by means of the advanced oxidation method was carried out in static conditions of laboratory reactors with capacity of 1 dm<sup>3</sup> equipped with magnetic stirrer. Chemical reagents were dosed on one-time basis in the beginning of each cycle, directly to the reactor. Reaction proceeded at pH = 3. In the experiment, the hydrogen peroxide dose was applied amounting to 3 g·dm<sup>-3</sup> at decreasing share of Fe<sup>2+</sup>. Three Fe<sup>2+</sup> : H<sub>2</sub>O<sub>2</sub> rates were examined: 1 : 10, 1 : 5 and 1 : 3. Effectiveness of oxidation of organic compounds versus time was controlled. Measurements were carried out: at the moment of reagent application; then after 1 min; 5 min; 30 min; 1 h; 1.5 h; and finally after 2 h.

In order to determine effectiveness of leachate treatment process from advanced oxidation chambers, concentration of organic substances was analyzed expressed as COD (determined by means of bichromate method) (HERMANOWICZ et al. 1999) the reaction value (HI 8818 pH-meter).

#### Effectiveness of treatment of leachates from municipal waste landfills

### **Biological methods**

It was found that in aerobic conditions, presence of packing had no effect on forms of nitrogen presents in reactor outflows. In reactors operating in anaerobicaerobic conditions, introduction of packing resulted in increase of ammonia nitrogen concentration and decrease of nitrates. Individual nitrogen forms occurring in leachates are presented in Figure 1.

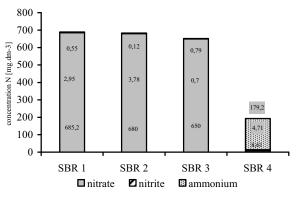


Fig. 1. Nitrogen forms in treated leachates

Introduction of packing resulted in losses of ammonia nitrogen, the fact being an indication of the effect of simultaneous nitrification and denitrification in active sludge.

Research work on leachates aimed at selection of optimum treatment technology is carried out since introduction of waste landfilling to the engineering practice.

High concentration of biomass and long age of sludge permit for more effective removal of organic substances with participation of slowly multiplying heterotrophic bacteria and maintenance of sufficient concentration of nitrification bacteria population. In order to increase biomass concentration and age of microorganisms, attempts are made to utilize highly efficient reactors such as fluidized beds or reactors with suspended biomass. Studies aimed at nitrification effectiveness improvement were also carried out with the use of combined methods — active sludge with biological membrane growing on a packing (movable or immovable carriers).

It follows from data published in the literature that packing materials may include: activated carbon, sand, plastics or PVC sponges. Organism grow on carrier surfaces or inside their porous structures (GIESEKE et al. 2002). To date, attention of researchers was focused only on increasing of the nitrification rate (VAN DE GRAAF et al. 1995). Simultaneous use of biomass carriers allows to improve yield of biomass and extend the period for which microorganisms of active sludge dwell in the reactor.

GIESEKE et al. (2002) in the SBR reactor packed with Kaldnes mouldings (SBBR) reached a decrease of ammonia nitrogen from 13 mg·dm<sup>-3</sup> to 0.8 mg·dm<sup>-3</sup>, with simultaneous increase of nitrates(V) up to 1.5 mg·dm<sup>-3</sup> at oxygen concentration in the reactor amounting to 5.4 mg·dm<sup>-3</sup>. After oxygen concentration being increased up to 6.3 mg·dm<sup>-3</sup>, the ammonia nitrogen level decreased from 40 mg·dm<sup>-3</sup> to 11 mg·dm<sup>-3</sup>, and content of nitrates(V) increased to 14 mg·dm<sup>-3</sup>.

It follows from studies of LOUKIDOU and ZOUDOULIS (2001) that it is possible to reduce ammonia nitrogen from leachates by 60%. The research work was carried out in a reactor packed with polyurethane cubes, and raw leachates were characterized with ammonia concentration at the level of 1,800 mg·dm<sup>-3</sup>. Low degree of reduction of ammonia nitrogen from leachates resulted from the fact that 18 h-long aeration period was insufficient to achieve full nitrification. Another reason can consists in presence of large amount of organic compounds prohibiting sufficient number of nitrification bacteria from multiplication.

ROSTRON et al. (2001) treated synthetic wastewater characterized with ammonia nitrogen concentration of 500 mg·dm<sup>-3</sup>. The research work was carried out in CSTRs under aerobic conditions. As packing, Linpor polyurethane cubes, Kaldnes polyethylene mouldings and capsules made of PVA (polyvinyl alcohol) were used. Full nitrification was achieved at the period of wastewater retention in reactor of 6; 3.4 and 2.2 d, respectively. Reduction of the retention time to 1.5 d resulted in occurrence of nitrites in the outflow. That could be caused by washing out *Nitrobacter* bacteria or too little amount of biomass with respect to the increasing load. At the retention time of 1 d, full nitrification in the reactor packed with Linpor and PVA blocks was achieved, while 90% nitrification was obtained with Kaldnes mouldings. After reduction of the retention time down 0.5 d, phase-II nitrification was observed. At the same time, 30% of ammonia nitrogen remained not removed.

In aerobic conditions, oxidation of ammonia nitrogen to nitrates(III) is carried out by *Nitrosomonas* sp. (phase I nitrification). Then, nitrite(III) nitrogen is oxidized to nitrate(V) nitrogen with the aid of *Nitrobacter* sp. (phase II nitrification) (VAN DER STAR et al. 2008).

Presently, a matter of significant importance for practical purposes related to removal of ammonia nitrogen from wastewater consists in processes of partial nitrification, oxidization of ammonia nitrogen in anaerobic (anoxic) conditions or combination of both processes. Among processes with dominant mechanism of ammonia nitrogen removal one can rate: partial nitrification and Sharon, Anammox and Canon methods (KHIN, ANNACHHARTE 2004).

Partial nitrification can constitute the first stage in a system with conventional denitrification or Anammox. In systems with partial nitrification, the denitrification process consists in reduction of nitrate(III) nitrogen to molecular nitrogen and occurs at higher rate compared to reduction of nitrate(V) nitrogen (TURK, MAVINIC 1989). In systems with partial nitrification and denitrification, reduction of demand for oxygen (by 25%) and organic carbon (by 40%) occurs. An additional advantage consists in lower biomass production and CO<sub>2</sub> emission (SCHMIDT et al. 2003, KHIN, ANNACHHARTE 2004).

Presently it is assumed that in reactors with full mixing, maintaining a short retention period (e.g. one day) and high temperature  $(30-40^{\circ}\text{C})$  is favorable for partial nitrification as it leads to washing out *Nitrobacter* sp. from the reactor. From the point of view of *Nitrobacter* sp. cultivation, low concentration of dissolved oxygen (less than 0.4 mg·dm<sup>-3</sup>) and high concentration of ammonia nitrogen (SCHMIDT et al. 2003) are unfavourable.

Modifications of Sharon process towards increase of its stability consist in shortening of period for which the sludge is retained in the reactor through abandonment of sludge recirculation. That way, instead of wastewater retention period, the sludge age is controlled that should be long enough in order to ensure sufficient multiplication of *Nitrosomonas* sp. in the active sludge chamber, but at the same time long enough to achieve complete washout of *Nitrobacter* sp. population from reactor (SCHMIDT et al. 2000). The final product of Sharon process is nitrate(III) nitrogen, but also not oxidized ammonia nitrogen. That is why Anammox method is recommended as the second stage for the purpose of complete ammonia nitrogen removal.

The Anammox process was discovered by MULDER and SCHMIDT et al. (1995). The authors examined anaerobic oxidization of ammonia nitrogen in presence of nitrate(V) nitrogen in the laboratory scale using an anaerobic fluidized reactor. Later research work carried out by VAN DE GRAAF, SCHMIDT et al. (1995) and BOCK, SCHMIDT et al. (1995) proved that rather nitrate(III) nitrogen, and not nitrate(V) nitrogen, is preferentially used as the electron acceptor. Nowadays it is a common assumption that Anammox consist in denitrification of nitrates(V) or (III) with ammonia nitrogen as the electron donor (KHIN, ANNACHHARTE 2004).

VAN DONGEN, SCHMIDT et al. (2001) examined removal of ammonia nitrogen in a two-stage system using the Sharon method as the 1<sup>st</sup> stage and Anammox as the 2<sup>nd</sup> stage treatment. At wastewater retention period amounting to 24 hours, they achieved a 53%-reduction conversion of ammonia nitrogen into nitrate(III) nitrogen in the 1<sup>st</sup> stage, while in the 2<sup>nd</sup> stage, at reactor load amounting to about 1.2 kg·m<sup>-</sup>  ${}^{3}$ ·d<sup>-1</sup>, the nitrogen removal efficiency exceeded 80%.

In the Canon process, elimination of nitrogen in a single reactor proceeds with participation of bacteria of genus *Nitrosomonas* sp. and plankton bacteria. In

conditions of limited concentration of dissolved oxygen, *Nitrosomonas* sp. oxidize ammonia nitrogen to nitrate(III) nitrogen, and plankton bacteria transform ammonia and nitrate(III) nitrogen to molecular nitrogen and, to some small concentration, to nitrates(V). In order to increase efficiency of the process and eliminate nitrate(V) nitrogen it is recommended to introduce inorganic substances as an external source of carbon (HAO, VAN LOOSDRECHT 2004).

#### **Chemical methods**

It was found that degradation of organic substances in leachates, at  $H_2O_2$  dose amounting to 3 g·dm<sup>-3</sup> and iron(II) doses equalling 1, 0.6 and 0.3 g·dm<sup>-3</sup>, efficiency of removal of organic compounds was high and amounted to 48% at Fe<sup>2+</sup> :  $H_2O_2$ ratio of 1 : 5 and 1 : 3 and 45% at Fe<sup>2+</sup> :  $H_2O_2 = 1 : 10$ . Variations of content of organic substances in time are presented in Figure 2.

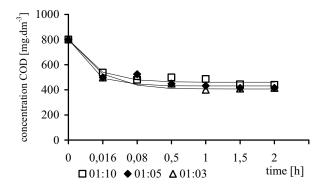


Fig. 2. Variations of content of organic substances in time at various  $Fe^{2+}$ : H<sub>2</sub>O<sub>2</sub> ratios

The best treatment results are achieved in two-stage systems representing a combination of biological and physicochemical methods. Physicochemical methods differ in efficiency, degree of complexity of technological solutions and related apparatus as well as in costs of individual processes. In the engineering practice, leachate treatment is commonly carried out by means of adsorption, coagulation/flocculation and reverse osmosis.

Degradation of refraction substances can be achieved by means of advanced oxidation. The method consists in generation of reactive hydroxyl radicals (OH<sup>•</sup>). Hydroxyl radicals (with oxidization potential of 2.8 V) react stronger than chemical oxidizers such as ozone or  $H_2O_2$  and are not selective, thus making possible for them to react with many chemical compounds.

It follows from review of the literature that leachate treatment can be quite effectively carried out with the use of Fenton's reagents ( $Fe^{2+}$  :  $H_2O_2$ ). In two-stage systems it can be applied both before and after biological treatment of leachates (LOPEZ et al. 2004; ZHANG et al. 2005).

The  $Fe^{2^+}$ :  $H_2O_2$  is of special importance, as it allows to avoid undesirable free radical reactions that may occur in case of excess of any of the reagents. When the proportion of  $Fe^{2^+}$  with respect to  $H_2O_2$  is optimal, OH radicals are used mainly for oxidization of organic substances (LOPEZ et al. 2004).

For that reason, many authors dealt with the problem of proper selection of proportion between reagents used for oxidation of organic substances. LOPEZ et al. (2004) demonstrated that at fixed  $H_2O_2$  dose amounting to 6.3 g·dm<sup>-3</sup>, COD in treated leachates decreased with increasing concentration of iron(II). After increase of the hydrogen peroxide dose up to 10 g·dm<sup>-3</sup> and at the proportion Fe<sup>2+</sup> :  $H_2O_2 = 1 : 8$ , the increase of BOD<sub>5</sub> : COD from 0.2 to more than 0.5 was observed. The authors concluded that final reaction products consist in short-chain organic acids resistant to further oxidation.

The advanced oxidation reaction is most effective al low reaction value. KANG, HWANG (2000) and LOPEZ et al. (2004) report that the highest efficiency of Fenton's reaction occurs at pH within the range 2.5-4. In the research work reported here, reaction of advanced oxidation with Fenton's reagent was carried out at pH = 3.

ZHANG et al. (2005) examined effectiveness of leachate treatment from municipal waste landfill in Sandtown characterized with organic substance concentration (COD) within the range 8,298–8,894 mg·dm<sup>-3</sup>. They demonstrated that at pH = 2.5, the course of Fenton's reaction was the most effective and rate of generation of iron (III) ions was the highest. The authors shown that with increasing temperature, efficiency of organic substance removal also increased. For instance, at initial COD level of 1,000 mg·dm<sup>-3</sup>, the efficiency increased from 42.3% (at temperature 13°C) to 56.2% (at 37°C), while at the initial level of 2,000 mg·dm<sup>-3</sup>, COD reduction increased from 31.6% to 44.8%, when temperature raised from 15°C to 35°C.

#### **Summary**

Usefulness of combining biological methods with chemical ones is beyond any doubt proved in case of removal of organic substances from leachates. In that situation, it is also important to ensure equally high nitrogen removal efficiency. In the opinion of ALBERS, KRÜCKEBERG (1992), KACZOREK et al. (2002) and ZHANG et al. (2005), even in cases when physicochemical processes such as ozonization or reverse osmosis are used, preliminary removal of nitrogen by means of biological methods is both appropriate and economically justified.

Research work on treatment of leachates from municipal waste landfill sites by means of the active sludge method in SBR reactors demonstrated that oxygen concentration reduction through introduction of an anaerobic phase into the SBR reactor operation and/or packing impairs the process of nitrification. Introduction of packing leads to increase of nitrogen losses and/or ammonia nitrogen concentration in effluents from reactors operating in anaerobic-aerobic conditions.

Further attempts to remove organic compounds from leachates with the use of Fenton's reagent proved that  $Fe^{2+}$ :  $H_2O_2$  ratio has insignificant effect on the

reaction rate and process effectiveness. Increase of  $H_2O_2$  in the mixture resulted in slight decrease of effectiveness of the process.

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