

ECOLOGICAL LIFE CYCLE ASSESSMENT OF MODIFIED NOVOLAKS WASTE USED IN INDUSTRIAL WASTEWATER TREATMENT

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Research article

Abstract: Ecological Life Cycle Assessment (LCA) applied in the assessment of the impact of products on the environment is a technique that allows for the evaluation of the environmental impact of polymeric flocculants used in industrial wastewater treatment. The possibility of conducting a full life cycle and thus manufacturing process analysis allows for reliable and accurate identification of the sources of environmental hazards and the impact of new products on the environment. Newly synthesized waste-based polymers are water soluble and possess the properties of flocculants, while reducing the parameters in industrial wastewater. In the paper, there are presented the results of the analysis conducted using LCA technique for the assessment of the impact of modified waste phenol formaldehyde resin (Novolak) on the environment. LCA technique was used to assess the impact of the new flocculant applied in the process of metallurgical wastewater treatment taking into account the environmental impact of the flocculant manufacturing process.

Keywords: LCA, modified Novolak waste, flocculants, metallurgical effluents treatment.

Introduction

Analysis of the current state LCA

Innovative technological solutions in industry are strictly connected with the acknowledged strategies of eco-development, and their preliminary aim is to limit environmental threats. According to the policy, Ecological Life Cycle Assessment of products may be the basis for planning and taking up activities in the local, national and global scale. Thanks to applying the method to assess production processes it is possible to study technological processes in the aspect of environmental safety.

The most renown assessment methods of production technology are: cumulated account, Schalteger&Sturm, BAT (*Best Available Technique*) and LCA (*Life Cycle Assessment*). LCA technique takes a special place within the concept of sustainable growth as an analytical tool, similarly

as substance flow analysis, risk analysis, product line analysis, environmental assessment in safety aspect (Kowalski et al, 2007; Kulczycka, 2011). Environmental risk analysis is especially important for ecological policy of countries (Bajdur et al., 2013; Sikorova and Bernatik, 2012).

The research on using LCA in the world is conducted in numerous scientific centres, as well as in industry. In the recent years, the experience in using this method has significantly increased both: in scientific centres and in industry. LCA is an environmental management technique, which allows to make an assessment of the impact on the environment of a product, process, sector of industry, sector of the economy, region, including the analysis of various environmental threats: physical, chemical and biological (Ščurek and Marsalek, 2011).

While conducting the research with the use of LCA one aims at considering all factors having impact on the environment, including the assessment

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of the relations between the input and output data created in each phase of the production process. Fig. 1 illustrates the relations between particular stages of the life cycle, starting from excavating the natural resources, through production and finishing with discharge or recycling of waste (Udo de Haes and Heijungs, 2007).

Complex attitude and the possibility to analyse complex and extended models of LCA technique results in a wide spectrum of usages. LCA technique may be combined with other tools of analysis and assessment, such as: SFA - Substance Flow Analysis

(Finnveden and Moberg, 2005), MFA -Material Flow Analysis (Udo de Haes and Heijungs, 2007), ERA- Environmental Risk Assessment (Bajdur and Miedzinska, 2006) or LCC - Life Cycle Cost (Kulczycka et al., 2003). For this purpose, computer software is used; in case of LCA the most commonly used software is Sima Pro. The software applies various calculation methods for European conditions.

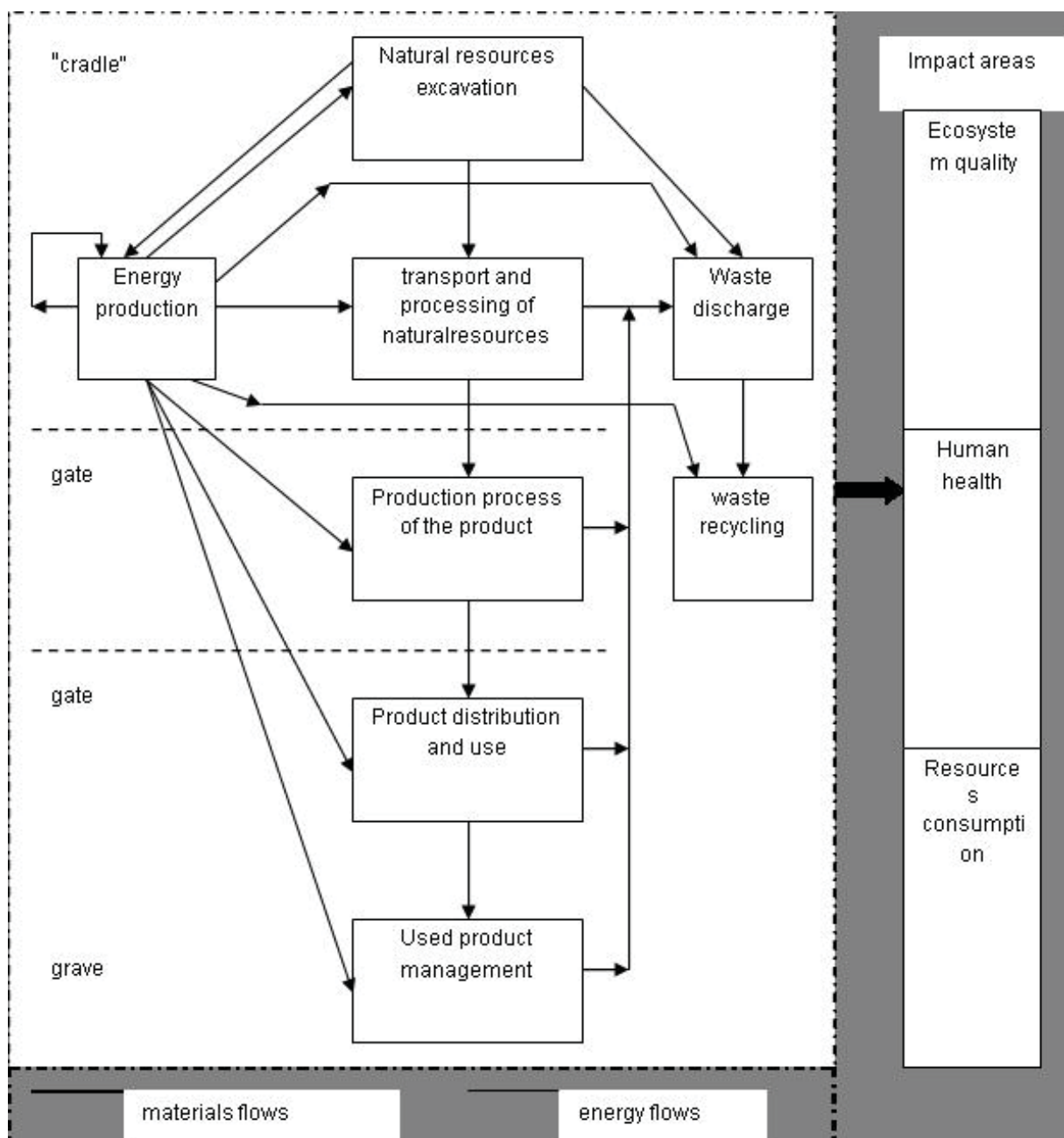


Fig. 1. Product life cycle diagram (Góralczyk and Kulczycka, 2001)

LCA and industrial wastewater management

The research and study of literature prove that Ecological Life Cycle Assessment may be used with good effect in the field of means and tools that improve industrial wastewater management. Using LCA to assess the results of inappropriate wastewater management may have good effect not only on the environment, but also on the economy by decreasing the expenditures on water and sewage management in industrial plants. In the recent years research in the environmental impact of different variants of wastewater treatment has been carried out. LCA is an acceptable technique and it contributes to solving technical problems connected with excessive creation of sewage and waste resulting from, among others, irrational waste management.

LCA is used in research on the development of new technologies of sanitary sewage and industrial wastewater. The aim is to increase ecological effect and reduction of risks in the existing technologies of wastewater treatment in order to achieve good chemical condition of water courses (Balkema et al., 2002; Thomas and McDougall, 2005; Bajdur and Henclik, 2009). Due to safety, before management plans of water basins are outlined a thorough analysis of the condition of water should be made. It is a priority to take up activities to reduce substances in wastewaters created in given sectors of industry and LCA may contribute to designing water management plans which would meet formal requirements. The requirement concerning water quality in metallurgical circuits results in the necessity to prepare the water constantly. In the waste water treatment processes in metallurgy (cooling circuits refreshment, coke oven gas purification, sinter, rolling, casting and galvanising processes) synthetic polyelectrolytes (floculants) are used. In the coagulation process coagulants are used (iron II salts and iron III salts and aluminium salts, which are supported with floculants of natural origin or synthetic ones (Bajdur and Henclik, 2008). Similarly, in galvanising plants mostly in automotive, agricultural machines, electronic, and electrochemical industry, in result of surface metal processing sewage is created, which, due to its toxicity, is one of the most dangerous and troublesome factors for the environment. These methods which allow for multiple use of baths are of the greatest importance. Thanks to these methods it is possible to regenerate galvanising bath and to recover ions of heavy metals from the used backwash waters. Thanks to them it is possible to decrease water consumption by closing the water circuit.

Chemical support of wastewater treatment is performed by using selected chemical reagents to act against the wastewater stream. Such reagents are: coagulants and floculants. In chemical treatment coagulation with e.g. iron sulphate, aluminium sulphate is used and polyelectrolytes are used as floculants. Polymer substances of a chain structure of the molecule, soluble in water are floculants and their molecular mass is from one million to several dozen millions of units. Floculants are used to speed up the sedimentation of grains, improve filtration, enrich minerals, modify the flotation process. One of the ways to obtain new generation polyelectrolytes may be a chemical modification of wastes from phenol-formaldehyde resins (Novolak). Recycling of this kind may be especially important in limiting the use of natural resources for the production of polyelectrolytes; additionally, it may be used to devise a new technology of producing floculants of a new generation (Bajdur, 2011, 2007, Bajdur and Sulkowski 2003a, 2003b, 2003c, 2005).

In this article the properties of amine derivatives of Novolak and sodium salts sulphone derivatives of Novolak are analysed. The products have been used as floculants in the treatment of industrial wastewater. The environmental impact assessment has been done of the new polyelectrolytes synthesis on the basis of waste from Novolak (phenol-formaldehyde resin) used to treat wastewaters from blast furnace coke oven gas. Life Cycle Assessment (LCA) was conducted using ReCiPe and IMPACT 2002+ methods with Sima Pro software.

Materials and methods

Materials for the research

The following substrates were used in the research: basic coagulant, floculants, commercial polyelectrolyte - *Praestol 2515*, metallurgical and galvanisation wastewater. The basic coagulant was the solution of aluminium sulphate (Al_2SO_4)₃ 18H₂O p.a. grade, which is often used to eliminate colloidal contaminants in process water and wastewater treatment. Commercial polyelectrolyte - *Praestol 2515* used in the comparative analysis of new generation polyelectrolytes and commonly used floculants on the basis of acrylamide is little anionic, of the density of 700 kg/m³ and viscosity 3000 m Pa.s. The floculant was used in the recommended pH range 3-8.

Polymers synthesised from Novolak wastes (phenol-formaldehyde resins) were used as flocculants to enhance the coagulation process of blast furnace gas treatment (metallurgical wastewater) The characteristics of new synthesised flocculants has been published in articles and monographs (Bajdur, 2011, 2013).

Tab. 1. New polyelectrolytes used in wastewater treatment (Balkema et al., 2002)

	Sodium salt of sulphate derivative of T Novolak waste	PS-N-T
	Amine derivative of SE Novolak waste	PA-N-SE

The research has been done using metallurgical wastewater (MW) coming from blast furnace gas treatment. While selecting the wastewater type the determining factor was the presence of cyanides, phenols, sulphates and chlorides in the wastewater. The wastewater was preliminary characterised by determining the selected physical and chemical indicators. Metallurgical wastewater was taken from the plant as momentary time samples and then they underwent sedimentation for 30 minutes in order to eliminate easily settling suspensions. Decanted wastewater was used in the technological study. Beforehand the physical and chemical properties had been determined by means of selected contamination indicators.

Research on metallurgical wastewater treatment

The research on metallurgical wastewater coagulation with the use of basic coagulant and newly synthesised flocculants was conducted according to the technological system diagrams (Fig. 1, 2) and the generally used research methodology of the coagulation process. In the research the coagulant alone or coagulant and flocculant were dosed, mixed and set apart for sedimentation. In each case the amount of sediment and sedimentation liquid were measured. Four samples of metallurgical wastewater were taken in the research on enhancing coagulation process with newly synthesised polyelectrolytes.

The description of the research carried out and the doses were done in previous studies and published in the monograph (Bajdur, 2011). Then the minimum dose of particular newly synthesised flocculants and commercial flocculant was determined also obtaining maximum decrease of turbidity. Similarly, as in the case of the minimum effective dose of the coagulant, also in respect of the flocculants the doses used in further research were sent in preliminary research (Bajdur, 2011).

On the basis of the results of the research it has been found that the optimum dose of the basic coagulant for metallurgical wastewater was 70 mg/dm³. Optimum doses of polyelectrolyte dose - sulphone derivative of Novolak T (PS-N-T)

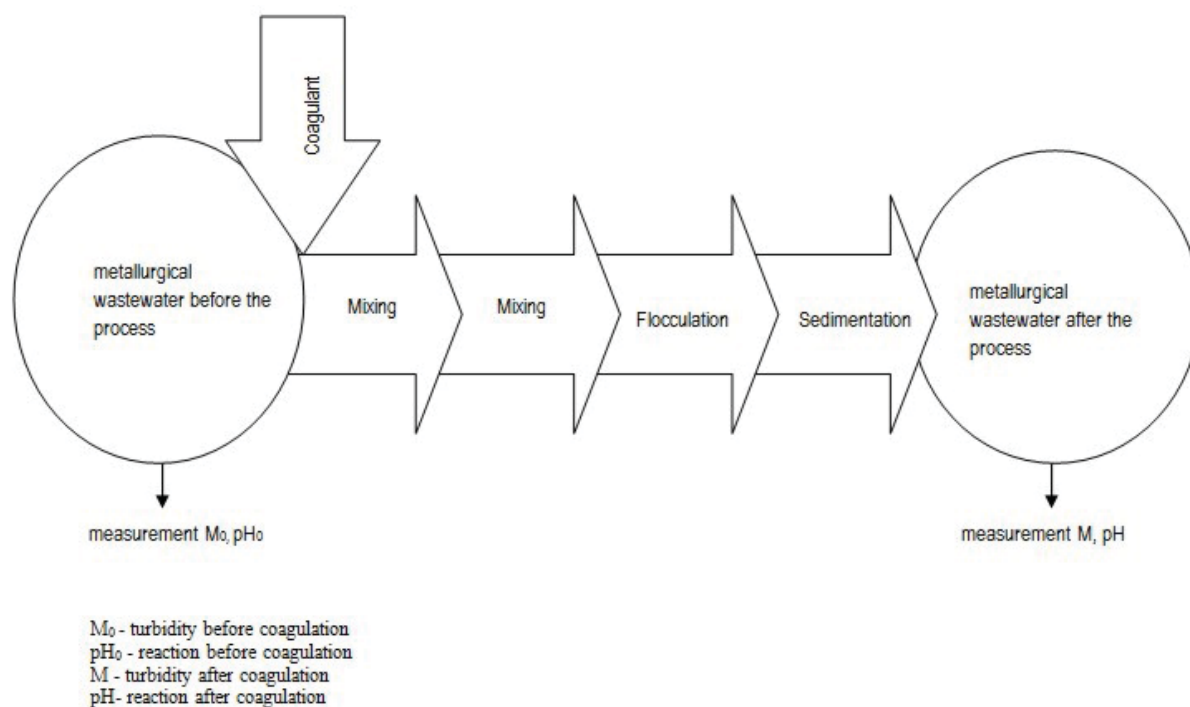


Fig. 2. Technological system diagram used in the research on coagulation of metallurgical wastewater with the use of coagulant (Bajdur, 2011)

and amine derivative of Novolak SE(PA-N-SE) were several dozen times smaller than the doses of aluminium sulphate. The doses in the range from 0.3 to 2.3 mg/dm³ were chosen for the research. Commercial polyelectrolyte Praestol 2515 was used in the following dose: 0.5 mg/dm³ and 1.0 mg/dm³ (Bajdur, 2013).

Turbidity was set in order to assess the efficiency of the process of coagulation and flocculation. Every measurement of turbidity was done seven times.

Turbidity was set using Turb 550 IR device providing quick and reliable measurement. The measurements were done using ISO 7027/DIN 27027 standard which is compliant with the recommendations of US EPA (Bajdur, 2011). The determination of the other physical and chemical indicators were done in certified laboratories, according to the current standards. Metallurgical wastewater of specified ranges of values of the indicators were selected for the research (Tab. 2).

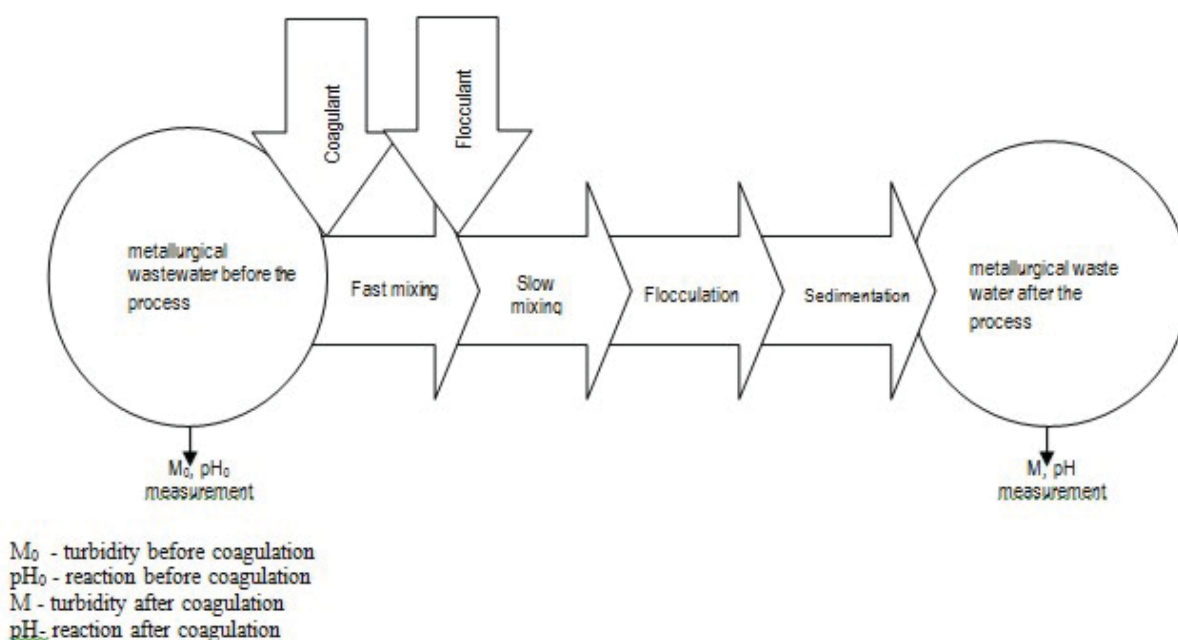


Fig. 3. Technological system diagram used in the research on coagulation of metallurgical wastewater with the use of coagulant and flocculant (Bajdur, 2011)

Tab. 2. Results of physical and chemical analyses of metallurgical wastewater

Indicator type	Unit	Range
Turbidity	NTU	160,0 ÷ 200,0
pH	-	6,80 ÷ 9,50
Phenoles	mg/dm ³	0,5 ÷ 5,5
Cyanides	mg/dm ³	0,5 ÷ 15,0
ChZT	mg O ₂ /dm ³	100,0 ÷ 300,0
Oxygen consumption	mg O ₂ /dm ³	20,0 ÷ 90,0
Ether extract	mg/dm ³	12,0 ÷ 35,0
Ammonia nitrogen	mg/dm ³	100,0 ÷ 290,0
Sulphates	mg SO ₄ /dm ³	130,0 ÷ 250,0
Chlorides	mg Cl/dm ³	800,0 ÷ 2000,0
General hardness	mg/dm ³	830,5 ÷ 1262,5
Dissolved substances	mg/dm ³	5,5 ÷ 5000,0
General suspension	mg/dm ³	15,0 ÷ 60,0

Tab. 3. Results of physical and chemical analyses of galvanising wastewater

Indicator type	Unit	Range
pH	-	7,8 ÷ 8,3
Turbidity	NTU	235,0 ÷ 276,0
Zn	mg Zn/dm ³	4,450 ÷ 7,450
Cu	mg Cu/dm ³	0,350 ÷ 0,455
Ni	mg Ni/dm ³	1,732 ÷ 1,923
Cd	mg Cd/dm ³	< 0,001
Pb	Mg Pb/dm ³	0,035 ÷ 0,056
Crog	mg Cr/dm ³	0,190 ÷ 0,450

Analogically, the process of coagulation of galvanising wastewater was done, according to the patterns presented in fig. 2 and 3.

The content of ions: Zn, Cu, Ni, Hg, Cd, Pb, Cr_{og} before and after the coagulation process supported by flocculants was determined according to the current standards.

The coagulation process was supported by using modified wastes from phenol-formaldehyde resins as flocculants: sodium salt sulphone derivative of Novolak T.

Assessment method of the impact of wastewater treatment with the use of flocculants on the environment

ReCiPe method used in the Ecological Life Cycle Assessment (LCA) aims at combining the methods used so far (mostly Eco-Indicator 99 and CML) and at determining their common frames. ReCiPe method joins intermediate points with the final ones. In IMPACT 2002+ method also combining two methods is proposed - CML and Eco-indicator 99 and similar categories of intermediate points and final points are grouped. Moreover, some categories of impact have been expanded. In the method described in the paper four categories of damage are proposed.

ReCiPe method was created by RIVM (*Rijksinstituut voor Volksgezondheid en Milieu* - National Institute of Public Health and Environment), CML (*Centrum voor Milieukunde Leiden* - The Institute of Environmental Sciences in Leiden), PRé Consultants, RUD (*Radboud Universiteit Nijmegen* - University in Nijmegen) and CE Delft. Its aim is to integrate the methods used so far and establishing their common frames (mostly the indicators specified in the publication *Handbook on LCA* and the commonly used method *Eco-Indicator 99*), as well as improving the quality and updating the knowledge on protection and condition of the environment. ReCiPe method joins the intermediate points with the final points, however it is recommended to use 18 intermediate points (impact categories).

ReCiPe and IMPACT 2002+ methods are a combination of two most renown assessment methods of life cycle: CML and Eco-Indicator 99. The difference between them is, among others the attitude towards the damage category "consumption of resources". In the IMPACT 2002+ method the amount of primal energy is the unit in MJ, however in ReCiPe method the increase in costs resulting from excavating resources (in dollars) (Kulczycka, 2011). Histograms have been devised using Sima Pro 8.1 software.

Results and discussion

Technological research

Analysis of the results of the research has shown that the turbidity of the examined wastewater was at a high level, up to 199.0 NTU in case of metallurgical waste and 276.0 NTU in case of galvanising waste. After using the basic coagulant, the value of turbidity in the examined wastewater decreased by over one hundred units NTU in comparison to the used commercial polyelectrolyte. However, pH valued decreased only in decimal or hundredth parts of the unit.

The analysis of the result of all the samples has shown that using the new synthesised polyelectrolytes made it possible to reduce the turbidity both: in metallurgical and galvanising wastewater over 60 % to almost 80 %, whereas using commercial polyelectrolyte caused the decrease of turbidity over 80 % using greater doses of commercial polyelectrolyte. The changes of turbidity of wastewater depended on the dosage and the optimum amount of the reagent oscillated in the range from 1.0 to 1.5 mg/dm³ of modified wastes from Novolaks. More effective doses of standard flocculant Praestol were 1.0 mg/dm³.

Selection of new flocculants (synthesised from the waste from Novolaks) for the research was preceded by thorough studies of the literature and experiments.

For the treatment of metallurgical waste an amine derivative of Novolak SE and sodium salt of the sulphone derivative of Novolak T were used, and for the reduction of heavy metals concentration in galvanising wastewater only sodium salt of the sulphone derivative of Novolak T was used, which resulted from previous experiments research. The analysis of the other indicators of wastewater and industrial waters conducted according to the current norms proved that using chemically modified polymer wastes in metallurgical wastewater-amine derivative of waste from phenol-formaldehyde resin (Novolak SE) and sulphone derivative of phenol-formaldehyde resin (Novolak T) leads to considerable decrease of almost all examined and analysed indicators (Tab. 4).

Using the flocculant PS-N-T and PA-N-SE in the coagulation process of metallurgical wastewater caused the decrease of chlorides concentration by approximately 90 %, and sulphates by approx. 60 %. Before the coagulation process metallurgical wastewater was characterised by the presence of organic compounds expressed by COD indicator

which was appropriately 210 mgO₂/dm³. In all the examined samples with the use of modified flocculants the drop of COD was greater than with the use of the commercial flocculant. The greatest effectiveness of eliminating organic compounds expressed by this indicator was obtained for PS-N-T flocculant (Tab. 4). And using PA-N-SE flocculant enabled to obtain slightly better effects in eliminating ether extract.

Other indicators were also significantly decreased. The greatest efficiency of eliminating organic compounds expressed by oxygen consumption was obtained after applying PS-N-T flocculant. It was at the level of 85.7 %. After applying PA-N-SE flocculant the efficiency was 60.8 %. General hardness was reduced by 80.9 % when PS-N-T flocculant was applied, and in case of

PS-N-SE flocculant similarly by 77.7 %. Also, the reduction of dissolved substances was considerable because the application of PS-N-T flocculant caused reduction by 93.4 %, and PA-N-SE flocculant by 89.8 %. By using the flocculants obtained from waste the amount of suspension in the wastewater decreased by 50 %. The analysis of the indicators has proven that new polyelectrolytes are more effective in comparison to commercial flocculant - *Praestol 2515*, apart from one parameter: suspension - in case of which the application of PS-N-T caused lower decrease of suspension than in the case of the used commercial polyelectrolyte. Reductions of cyanides and phenols (below 0.005 mg/dm³) in the examined wastewater were of particular importance.

Tab. 4. The results of the analysis of metallurgical wastewater after coagulation process enhanced by selected flocculants - amine derivatives of waste from Novolaks and standard polyelectrolyte

Indicator type	Indicator value before treatment	Indicator value after treatment PS-N-T	Indicator value after treatment PA-N-SE	Indicator value after treatment P-2515
Phenols [mg/dm ³]	4,9	< 0,005	< 0,005	< 0,005
Cyanides [mg/dm ³]	2,3	< 0,005	< 0,005	< 0,005
COD [mg O ₂ /dm ³]	210	42,6	71,5	95,7
Oxygen consumption [mg O ₂ /dm ³]	53,0	7,6	20,8	30,2
Ether extract [mg/dm ³]	13,9	9,5	9,1	11,3
Ammonia nitrogen [mg/dm ³]	161,0	11,20	11,96	82,78
Sulphates [mg SO ₄ /dm ³]	150,4	80,3	97,5	140,7
Chlorides [mg Cl/dm ³]	1556,5	146,7	148,9	1350,3
General hardness [mval/dm ³]	1111,0	212,1	247,5	858,5
Dissolved substances [mg/dm ³]	4791,3	316,0	489,0	2954,3
General suspension [mg/dm ³]	28,9	14,6	12,9	11,9

Table 5. The results of the analysis of the content of heavy metal ions in galvanising wastewater after the coagulation process enhanced by sodium salt of sulphone derivative of Novolak T

Indicator type	Unit	Value before wastewater treatment	Value after wastewater treatment
pH	-	7,8	7,5
Turbidity	NTU	276,0	85,8
Zn	mg Zn/dm ³	7,54	2,64
Cu	mg Cu/dm ³	0,48	0,23
Ni	mg Ni/dm ³	1,92	0,56
Cd	mg Cd/dm ³	< 0,003	< 0,001
Pb	mg Pb/dm ³	0,04	0,02
Cr _{og}	Mg Cr/dm ³	0,35	0,07

Also, an examination of galvanising wastewater after coagulation process enhanced by sodium salt of sulphone derivative of Novolak T (Tab. 5) was done. Sulphone derivative of Novolak was used in the concentration of 0.01 mg/dm³ after using hydrated aluminium sulphate in the concentration of 20 mg/dm³.

The analysis of the results of the examined sample showed considerable decrease of the concentration of hard metals ions in galvanising wastewater after using the new type flocculant.

Ecological Life Cycle Assessment(LCA)of new type flocculants

Life Cycle Assessment of amine derivative of Novolak SE and sodium salt sulphone derivative

of Novolak T comprises of the assessment of potential production process (Bajdur, 2011) and the assessment of the impact of the process of supporting the coagulation of metallurgical waste with new synthesised polyelectrolytes on the environment. The assessment has been done according to the principles of the methodology. It takes into consideration the impact of the production of the examined flocculants on the environment. In the inventory tables for wastewater treatment process include such data as: amounts of treated wastewater, amount of flocculants PS-N-T and PA-N-SE selected for the research and the make-up water used to prepare them, as well as electrical energy consumption. In the input and output data set also the reduction of the indicators in the examined wastewater presented in Tab. 4 was considered. On the basis of the inventory tables for metallurgical waste treatment using ReCiPe and IMPACT 2002+ method the results of the analysis are presented in the form of normalisation histograms. The impact on the environment of treatment of a particular amount of metallurgical wastewater is presented. The daily amount of treated metallurgical wastewater is taken for the functional unit, that is 20 000 m³ using the required amounts of flocculants for the examined wastewater. The comparative normalisation histograms of the metallurgical wastewater treatment with the use of the new type of flocculants PS-N-T and PA-N-SE show in both ReCiPe methods the positive effect of sodium salt sulphone derivative of Novolak T (PS-N-T) (Fig. 4, 5).

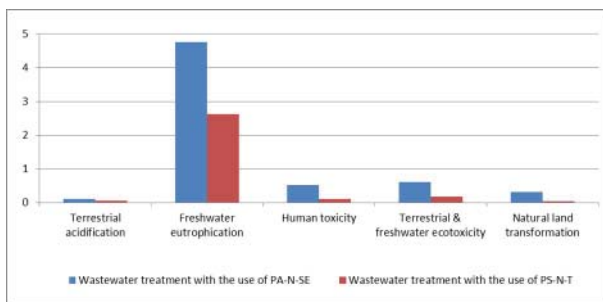


Fig. 4. Normalisation histogram for metallurgical wastewater treatment using new type flocculants PA-N-SE and PS-N-T according to ReCiPe method

The negative impact of amine derivative of Novolak SE on the environment is particularly clear according to ReCiPe method in the following categories: water eutrophication, human toxicity, ecotoxicity (freshwater and land, as well as land development (increasing values on the vertical axle, that is increasing potential impact). Similarly, in IMPACT 2002+ method, the negative impact of PA-N-SE is considerable and it concerns mostly

non-organic compounds on the respiratory system, global warming and non-renewable energy.

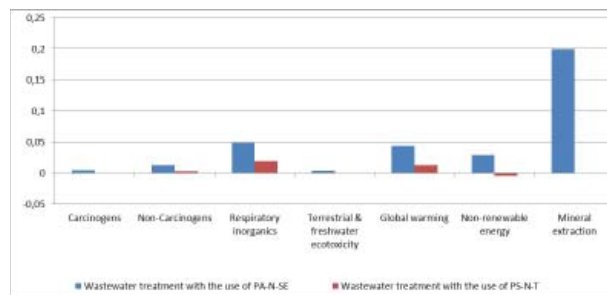


Fig. 5. Normalisation histogram for metallurgical wastewater treatment using new type flocculants PA-N-SE and PS-N-T according to IMPACT 2002+ method

The environment assessment of the process of enhancement of metallurgical wastewater coagulation with new synthesised polyelectrolytes was conducted according to the principles of the methodology.

Conclusions

Ecological Life Cycle Assessment (LCA) shows that the flocculants obtained from waste not only are effective, but also, they have less load on the environment in comparison to the commercial product used so far. This is very important from the point of view of the impact of industrial plants on the environment (Bartkiewicz, 2000). Industrial plants implement environment management programmes introducing the best accessible techniques (Niesler and Lackowski, eds, 2005). This applies also to water and sewage management. Rational water management is most of all using closed water circuits and this is possible thanks to using new, effective methods or means used in wastewater treatment. Synthesis and using new flocculants of waste from phenol-formaldehyde resins (Novolak T and SE) makes it possible to on the one hand limit waste in the environment, and the other hand to get good effectiveness of the flocculation process. The treated wastewater was characterised not only by lower turbidity in comparison to the turbidity of the wastewater with the use of commercial preparation. Due to the costs, the amount of applied chemical reagents for the coagulation process is of great importance. Similar effectiveness of removing contaminants was obtained while using several times smaller doses of modified preparations (amine derivative of Novolak SE and sulphone derivative of Novolak T) in comparison to commercial product. Among newly synthesized PS-N-T and PA-N-SE

flocculants used in supporting the metallurgical wastewater coagulation process PS-N-T flocculant indicated greater effects for reduction in pollution indicators. To the greatest extent, there were reduced the pollution indicators: oxygen consumption, ammonia nitrogen, sulphates, chlorides, general hardness.

Life Cycle Assessment of newly synthesized flocculants indicated that the acquisition of flocculants from waste phenol formaldehyde resin is highly beneficial for the environment due to high toxicity of waste resins which produce phenol under the influence of physical factors.

There was conducted the analysis of the research results presented graphically in figures created with Sima Pro software using, included in the tables, the values obtained by the basic research of treated wastewater using a new generation of flocculants. Less environmentally friendly flocculant in

metallurgical wastewater treatment was a derivative of PA-N-SE Novolak in both applied research methods.

The conducted LCA shows that the flocculants obtained from waste are not only effective, but also, they load the environment to lesser extent, considering most of all the balance of materials and energy. The streams of treated wastewater may become the source of water used for particular purposes plants.

The research on industrial wastewater treatment technologies is the field where LCA may be successively used in order to increase the ecological effect. Similarly, as in the case of research on waste disposal with the use of LCA, also alternative scenarios of sanitary sewage treatment technologies and using sediments, which show different solutions including the principles of sustainable growth are of considerable importance.

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