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Positive examples of wastewater treatment effectiveness in "Natron-Hayat" Maglaj factory

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A B S T R A C T

In the paper are described the basic characteristics of wastewaters in the cellulose and paper factory "Natron-Hayat" Maglaj. Particular emphasis is placed on the description of the technological process of wastewater treatment at the "Natron-Hayat" Maglaj factory which is represented in the paper by a technological scheme and represents one of the more complex systems for wastewater treatment. In the experimental part, the results of the efficiency of the wastewater treatment system at the "Natron-Hayat" Maglaj factory were given. It can be said that the basic raw materials for paper production are cellulose fibers and water. The role that water has in the cellulose and paper industry as the solvent, agent for cooking and washing of the produced pulp, indicates the presence of water in almost all stages of cellulose fiber production (Žarković et al., 2004). In accordance with the principles of sustainable development, cellulose and paper industry must face strict regulations on the protection of the environment which includes rationalization of raw material consumption, water, energy and chemicals, with minimal negative environmental impact. Wastewaters generated in the factory "Natron-Hayat" Maglaj are subjected to the purification before discharging into surface water recipient. The wastewaters from "Natron-Hayat" Maglaj factory are discharged into river Bosna.

1. Introduction

The term wastewater in the broad sense refers to any water which has changed its physical, chemical and biological characteristics to a greater or lesser extent while using, or it can be said one used water is wastewater (Popović, 2001). The role of water in the cellulose and paper industry is so important that it is often considered to be another source of raw material in addition to plant species. It was interesting, but also useful to examine the quality of the wastewater as well as the efficiency of the purification system, since the wastewater from the cellulose and paper industry is highly polluted and it is discharged into the river Bosna, which goes further directly through the town of Maglaj affecting the plants and living beings in the mentioned river, and thus influences the life quality of citizens.

The largest volume of water in the cellulose and paper industry is used on defibring of cellulose raw materials and the production of suspension, whereby the cellulose and paper industry is one of the largest consumers and natural resource polluters (Žarković et al., 2004). Large amounts of water are used during the process of washing pulp which is very important both from an economic and from an environmental point of view, because the quality of the pulp washing is directly related to the efficiency of chemicals regeneration and the quality of generated wastewaters. The purpose of the cellulose fiber washing process is to economically remove the maximum of soluble organic and inorganic materials with a minimum of fresh and recycled water (Halilović, 2016).

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In the domestic and foreign literature (Žarković et al., 2004; Popović, 2001; Gačeša and Klašnja, 1994), numerous examples can be found as solutions to the problem of water and wastewater quality in the cellulose and paper industry. The end objective which is rational water consumption and minimal impact on natural water courses, according to previously made researches can be achieved by increasing the degree of closed circular flow of water and the application of modern methods for wastewater treatment. Rational use of water with optimum recirculation level provides less load of central wastewater treatment plant (Žarković et al., 2004).

Direct discharge of wastewater, without prior treatment, to a nearby river, lake or sea, is the cheapest and easiest way, but for the environment it is the most unfavorable solution. One of the measures that the company establishes in addition to obligatory wastewater treatment is the imposition of increasingly stringent requirements for the more quality effluent, so that it can be reused, even for technical purposes (Popović, 2001).

The task of wastewater purification is to remove wastewater pollution to the extent that treated wastewater can be discharged into the recipient without any harmful consequences (Gačeša and Klašnja, 1994). Thus, the primary purpose of wastewater treatment is to provide a more complete release from various pollutants and toxics, whose dimensions are within very wide ranges, from simple junctions to large, floating pieces of material. The degree to which wastewater should be purified depends on its composition, mass, class and recipient size and legal regulations. Most countries in the world, in their legislation, require such water quality that will be discharged in the recipient that is necessary previously subject it to treatment (Đuković et al., 2000).

Wastewaters generated in the factory "Natron-Hayat" Maglaj are subjected to the purification before discharging into surface water recipient. The aim of this work is to examine the efficiency of devices for wastewater treatment in the factory "Natron-Hayat" Maglaj and to examine the quality of purified wastewater. Results are expressed through the reduction degree of the following parameters: chemical oxygen demand (COD), suspended matter content (SM) and biological oxygen demand (BOD_5) of treated wastewater being discharged into a watercourse.

2. Characteristics of wastewaters in the cellulose and paper industry

Wastewaters from cellulose and paper industry are loaded with organic pollution and contain chemicals that are used in the paper production process, small pieces of wood, cellulose fiber, which if not removed can cling to the gills of fish, dissolved lignin from wood, sulfur compounds. They contain a large amount of solid substances, which if they are not purified and discharged into the rivers quickly cover the bottom of the river destroying the fish and aquatic world which depends on the food from the bottom of the river (Bobar and Bajramović, 2011). There are black and white wastewaters and their characteristics are given in table 1. Black wastewaters originate from the cellulose production process, and the largest quantities are formed after washing the cellulose and they are dark brown in color. Their coloration comes from the presence of lignin, and this color affects photosynthesis ability of the aquatic plant (Bobar and Bajramović, 2011). Wastewater from the paper sector is called white water.

These wastewaters can have a variable character, both in quantity and quality. The amount of wastewater can vary widely, both during a day and for a longer period of time. Especially it is emphasized impact discharge of wastewater, that indicates the discharge of a larger amount of wastewater in a relatively short time.

Biological oxygen demand and chemical oxygen demand are the most common criteria for wastewater pollution. The pollutant criteria serve to estimate the damage caused by untreated wastewater to the recipient and to select the treatment method. For the sizing and evaluation of the purification plant performance, it is not only necessary to know the concentration but also the amount of pollution (load) that is being introduced. Load is expressed through the volume of wastewater that reaches the plant in the unit of time (Gačeša and Klašnja, 1994).

3. Wastewater treatment in "Natron-Hayat" Maglaj factory

Methods of wastewater treatment can be classified in various ways. Wastewater treatment is usually divided into (Gačeša and Klašnja, 1994):

- previous processing,
- primary,
- secondary,
- tertiary purification (in some industries), and
- processing and disposal of sludge generated during wastewater treatment.

Previous treatment of wastewaters involves removal of rough suspended and floating material and removal of inert material. Primary purification removes suspended and emulsified matter from wastewater by precipitation. Secondary purification removes the colloidal and part of the dissolved organic matter by chemical or biological means. Tertiary purification is used to remove residual pollution and for disinfection of purified water (Gačeša and Klašnja, 1994).

Figure 1 shows the technological process of wastewater purification in the "Natron-Hayat" factory.

In the equalization tank pH value of wastewater is measured, pH correction is done and coagulant dosage of $Al_2(SO_4)_3$ is added. The role of the coagulant is to neutralize the negatively charged particles and then to adsorbe them to itself thus creating conditions for their faster precipitation (Bušatlić et al., 2013). Then the wastewater through the pump is transferred from the egalitization tank to the primary clarifier tank. In the primary clarifier that is radial, it is added as flocculant

Table 1
Cellulose and paper industry wastewater characteristics (Mečević, 2009)

Characteristics	Black wastewaters	White wastewaters	All wastewaters
Amount	18,000 m^3/day	22,000 m^3/day	40,000 m^3/day
BOD_5	7,000-12,000 kg/day	3,000-6,000 kg/day	10,000-18,000 kg/day
COD	14,000-35,000 kg/day	6,000-13,000 kg/day	20,000-48,000 kg/day
SM	3,500-7,000 kg/day	6,000-13,000 kg/day	9,500-20,000 kg/day
Max BOD_5	700 mg/dm^3	350 mg/dm^3	
Max COD	2,000 mg/dm^3	750 mg/dm^3	
Max SM	400 mg/dm^3	750 mg/dm^3	

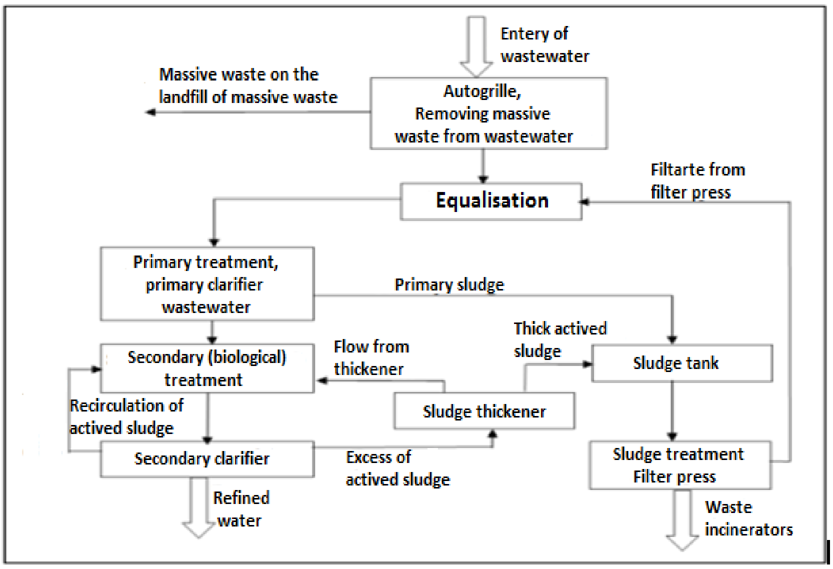

Figure 1. Schematic view of wastewater purification in "Natron-Hayat" Maglaj factory (Mečević, 2009)

an anionic polyelectrolyte. Precipitated particles from the primary clarifier are removed as a thickened slurry called "Primary Sludge". Primary sludge is drained by means of two submersible pumps which are placed on the scraper sludge and they work periodically. In the channel between the primary clarifier tank and the first aeration pool nutrients, iron sulfate ($FeSO_4$) and sodium hypochlorite ($NaClO_4$) are added. There are three aeration tanks for (secondary) biological treatment and one secondary clarifier divided into three sections. In aeration tanks is used activated sludge that contains microorganisms that will degrade organic pollution. The air is constantly introduced with a diffuser.

The primary sludge is taken directly to the sludge reservoir, while the sludge generated in the secondary purification (the active sludge) partly returns to the first aeration pool and partly goes to the sludge thickener and then into the sludge reservoir. The mixed primary sludge and the excess of the active sludge are sent to the trap filter press where dehydration of sludge is performed. In order to achieve better dehydration effects before the filter press the mixed sludge is treated with a cationic polyelectrolyte. The cake obtained on the filter press is then sent to the burning boiler and the filter through the sewerage network is sent to an equalization pool. Wastewaters from all parts of the plant are collected through the collecting channel and then treated at wastewater treatment plant (Halilović, 2016).

4. Experimental part

Experimental studies have been conducted in the period from 05.10.2015. until 29.04.2016., with the exception of 03.03.2016. until 04.04.2016. when factory remounting was done, in cellulose and paper factory "Natron-Hayat" Maglaj.

To determine the efficiency of the system for wastewater treatment during a period of a six months, every work day the following samples were taken:

- water samples after primary processing, ie. primary sedimentation and
- water samples at the entrance and the exit of the purification system, and the following parameters are analyzed:

  - the chemical oxygen demand (COD),
  - the biological oxygen demand (BOD_5), and
  - the content of suspended matter (solids) (SM).

The regulation on conditions for discharging industrial wastewater into the environment Federation

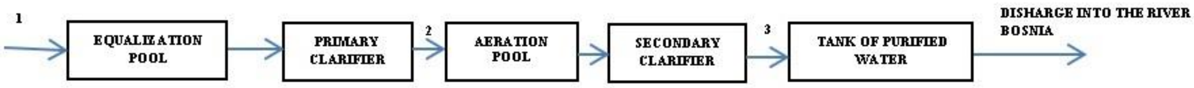

Figure 2. Schematic view of water sampling places: 1-sample taking and analysis of unpurified water: COD, BOD_5 and SM; 2-sampling after primary clarifier and COD and SM analysis; 3-purified water sampling and determination of COD, BOD_5 and SM (Halilović, 2016)

a) b) c) d)

Figure 3. The sampling places of wastewater: a) at the entrance to the treatment plant, b) after the primary clarifier, c) the overflow from the secondary clarifier-purified water, d) the output of treated wastewater into the river Bosna (Halilović, 2016)

of BiH determines and prescribes emission limit values for wastewater discharging into the environment and minimal percentage of load reduction compared to the parameters COD, BOD5 and suspended solids content after the secondary purification. The effectiveness of the devices for wastewater treatment was analyzed after:

- the primary purification (primary treatment or precipitation) and
- the secondary (biological) purification (after which purified water is discharged into the river Bosna).

4.1. Sampling

Figure 2 shows a schematic view of the places where the sampling of wastewater was done.

Figure 3 shows the sampling places in the wastewater treatment section.

4.2. Analyzed parameters and examination methods

Water sampling is done so that the water from water sampling place is first stired, and then from the various parts and depths a certain volume of water is poured in the bottle.

COD is chemical oxygen quantity demanded for organic compounds oxidation and part of inorganic salts, and it is expressed as mg/dm^3 O_2. COD was examined according to the internal method of "Natron-Hayat" Maglaj, and is based on heating a sample at boiling temperature with a strongly acidic solution of dichromate in the presence of a catalyst Ag_2SO_4 (Halilović, 2016). The dichromate is added to the excess and the unused portion is determined by titration with a standard solution of ferro ammonium sulfate ($Fe(NH_4)_2(SO_4)_2 \cdot 6H_2O$). The amount of dichromate consumed is calculated as the equivalent of the oxygen intake. Dichromate is reduced to equation (Rajaković-Ognjanović, 2016):

$$Cr_2O_7^{2-} + 14H^+ + 6e^- \rightarrow 2Cr^{3+} + 7H_2O \quad (1)$$

$HgSO_4$ is added, which prevents oxidation of chloride ion into chloride.

BOD is the amount of oxygen necessary to the water sample microorganisms under aerobic conditions at a temperature of 20 °C for a certain time period of incubation, to oxidize the organic matter in water (Gačeša and Klašnja, 1994).

Biological oxidation of organic matter is a very slow process and theoretically it would take an infinite amount of time to complete it. However, for a period of 20 days, 95 to 99 % of the original organic matter is oxidized. Since, from a practical point of view, that is a very long period of time, it was adopted that the incubation period for standard BOD determination is 5 days, at which time, at a temperature of 20 °C, it is oxidised 60-70 % of originally present organic matter (BOD_5) (Gačeša and Klašnja, 1994). The first stage in which it happens carbon oxidation to CO_2 and hydrogen oxidation to H_2O lasts relatively short, 7 to 10 days. The second stage in which nitrogen is oxidized to nitrite and then to nitrate, nitrification, lasts considerably longer. As water temperature rises, oxygen consumption and oxygen oxidation increase. The BOD is also the basic indicator that serves as an indicator of the impacts of wastewater on the receiver water where the oxygen content is reduced. As a rule applies, that during the determination of the degree of purification of polluted water at the plants it is necessary to achieve in the effluent such a BOD value that will not reduce the dissolved oxygen content downstream in the watercourse. For determination of BOD_5 manometer method and device BOD SYSTEM Oxidirect, Lovibond, shown in Figure 4 is used

Figure 4. Device for BOD_5 determination, BOD OXIDIRECT, LOVIBOND (BOD Measurement System, 2016).

Figure 5. Procedure of suspended matter examination – internal method "Natron-Hayat" Maglaj (Halilović, 2016)

according to manual that comes with the instrument (Halilović, 2016).

Suspended substances in water are 70 % organic and 30 % inorganic. They pollute the water esthetically and ecologically. If they are mainly of organic origin, their subsequent chemical degradation changes the composition of water. They precipitate in calm waters and endanger the living beings at the bottom of the water, and also reduce the transparency of the water.

Suspended solids were determined according to the internal method "Natron-Hayat" Maglaj (figure 5) (Halilović, 2016):

$$M_1 = \frac{(m_1 - m_0) \cdot 1000}{V} \qquad (2)$$

M_1 – content of suspended matter (SM), (mg/dm^3),
m_0 – filter paper mass, (mg),
m_1 – filter paper mass after filtration and drying, (mg), and
V – sample volume, (cm^3).

According to the regulation on conditions for discharging wastewater into the environment and the public sewage system of Federation of BiH the limit value of parameter COD of the wastewater that are discharged into surface water recipients is 125 mg/dm^3 O_2, of parameter BOD_5 is 25 mg/dm^3 O_2 and total suspended solids is 35 mg/dm^3 (Law on Water, 2006).

5. Results and discussion of the results

Given the large number of obtained data in the tables average values for individual months of the examined parameters are shown. Table 2 shows the COD values for unpurified and purified wastewater (after secondary treatment) and the wastewater after the primary sedimentation (p.s.). Also, in the table COD parameter reduction degree after the primary treatment (p.t.) and after secondary purification (s.p.) is given.

COD parameter reduction degree or purification efficiency (P.E.) compared to the COD parameter after the primary treatment is determined as follows:

$$\text{P.E. (COD)}_1 = \frac{913{,}343 - 417{,}234}{913{,}343} = 0.543 = 54.3\ \%$$

The average degree of reduction after primary precipitation is 54.3 %. After the water enters the equalization tank, it is dosed, among other chemicals, $Al_2(SO_4)_3$, that serves as a coagulant, which will reduce the organic and inorganic loads. Then, during the primary precipitation, a certain amount of flocculant is added in the wastewater, which will allow faster and more efficient precipitation of the impurities contained in the water. Based on a given reduction degree, it can be concluded that a large part of organic water pollution is lost in primary clarifier.

From Table 2 it can be seen that the COD average value before purification was 913,343 mg/dm^3 O_2 and after the secondary purification 111,014 mg/dm^3 O_2. On the basis of these two values can be determined COD parameter reduction degree or percentage of load reduction compared to the parameter COD after secondary purification:

$$\text{P.E. (COD)}_2 = \frac{913{,}343 - 111{,}014}{913{,}343} = 0.878 = 87.8\ \%$$

Table 2
The average efficiency of COD values decrease after primary treatment and secondary purification for each month in the examination period (Halilović, 2016)

Month	$COD_{before\ treatment}$ (mg/dm^3 O_2)	$COD_{after\ p.s.}$ (mg/dm^3 O_2)	$COD_{after\ treatment}$ (mg/dm^3 O_2)	Reduction degree of $COD_{after\ p.t.}$ (%)	Reduction degree of $COD_{after\ s.p.}$ (%)
October	867,513	374,421	108,205	56.8	87.5
November	949,306	401,667	129,667	57.7	86.3
December	927,200	448,975	128,300	51.6	86.2
January	971,265	447,382	116,088	53.9	88.0
February	881,222	399,556	91,111	54.7	89.7
April	883,550	431,405	92,715	51.2	89.5
average	913,343	417,234	111,014	54.3	87.8
Permitted value of COD reduction degree after the secondary water treatment by Regulation of Federation of BiH					**75 %**

Table 3
The average efficiency of SM values decrease after primary treatment and secondary purification for each month in the examination period (Halilović, 2016)

Month	$SM_{before\ treatment}$ (mg/dm^3)	$SM_{after\ p.s.}$ (mg/dm^3)	$SM_{after\ treatment}$ (mg/dm^3)	Reduction degree of $SM_{after\ p.t.}$ (%)	Reduction degree of $SM_{after\ s.p.}$ (%)
October	498,250	53,600	10,600	89.2	97.9
November	767,444	64,444	15,000	91.6	98.0
December	643,600	50,200	17,200	92.2	97.3
January	577,176	54,412	12,471	90.6	97.8
February	638,944	48,222	11,889	92.5	98.1
April	569,500	54,400	13,100	90.4	97.7
average	615,819	54,213	13,377	91.2	97.8
Permitted value of SM reduction degree after the secondary water treatment by Regulation of Federation of BiH					**90%**

Regulation on the conditions for discharge of wastewater into the environment of Federation of BiH defines the minimum percentage of the load reduction in relation to the COD parameter, mg/dm^3 O_2, which must reach the plant for wastewater treatment (after the secondary water treatment, and according to the mentioned regulation secondary treatment means second level of purification which generally includes biological treatment with a secondary settlement), and amounts 75 %. Based on the analyzed data can be concluded that the COD percentage reduction, mg/dm^3 O_2 of the plant for wastewater treatment in a factory "Natron-Hayat" Maglaj is very good.

Table 3 shows the suspended matter content values for unpurified and purified wastewater (after secondary treatment) and the wastewater after the primary sedimentation. Also, in the table SM parameter reduction degree after the primary treatment and after secondary purification is given.

SM parameter reduction degree in the wastewater or purification efficiency (P.E.) compared to the SM parameter after the primary treatment is determined as follows:

$$\text{P.E. (SM)}_1 = \frac{615{,}819-54{,}213}{615{,}819} = 0.912 = 91.2\ \%$$

Based on the data from Table 3 it can be concluded that the efficiency of the primary clarifier is very good and most of the suspended particles are precipitated in the primary sedimentation tanks.

From Table 3 it can be seen that the SM average value before purification was 615,819 mg/dm^3, and after the secondary purification 13,377 mg/dm^3. SM parameter reduction degree or percentage of load reduction compared to the SM parameter after secondary purification:

$$\text{P.E.(SM)}_2 = \frac{615{,}819-13{,}377}{615{,}819} = 0.978 = 97.8\ \%$$

The degree of suspended matter reduction of unpurified water, after its purification in wastewater treatment plant is 97.8 %. Regulation on the conditions for discharge of wastewater into the environment of Federation of BiH defines the minimum percentage of the load reduction in relation to the SM parameter, mg/dm^3, which must reach the plant for wastewater treatment after the secondary water treatment and amounts 90 %.

In table 4 is shown the average efficiency of BOD_5 values decrease after secondary purification.

The efficiency of purification devices for the wastewater treatment in relation to BOD_5 parameter is:

$$\text{P.E. (BOD}_5) = \frac{180{,}124-12{,}642}{180{,}124} = 0.93 = 93.0\ \%.$$

Table 4
The average efficiency of BOD_5 values decrease after secondary purification for each month in the examination period (Halilović, 2016)

Month	$BOD_{5\ before\ treatment}$ (mg/dm^3 O_2)	$BOD_{5\ after\ treatment}$ (mg/dm^3 O_2)	Reduction degree of $BOD_{5\ after\ s.p.}$ (%)
October	145,000	15,000	89.7
November	173,143	19,750	88.6
December	184,500	9,600	94.8
January	189,625	10,250	94.6
February	196,143	10,500	94.6
April	192,333	10,750	94.4
average	180,124	12,642	93.00
Permitted value of BOD_5 reduction degree after the secondary water treatment by Regulation of Federation of BiH			**70-90%**

6. Conclusion

The effectiveness of devices for wastewater treatment in the factory "Natron-Hayat" Maglaj fully meets the legal regulation on conditions for discharging wastewater into the environment. Results are expressed through the reduction degree of the following parameters: COD, suspended matter and BOD_5 of the purified wastewater being discharged into a watercourse. The obtained values of the reduction degree of examined parameters are significantly higher than the legally prescribed minimum values of Federation of BiH. The average value of COD parameter for purified wastewater discharged into the surface water recipient is 111,014 mg/dm^3 O_2, the average value of suspended substances amounts to 13,377 mg/dm^3 and the average value of BOD_5 is 12,642 mg/dm^3 O_2. All the values are lower than those prescribed by the Regulation. This paper confirms that the quality of wastewater treatment in the cellulose and paper factory "Natron-Hayat" Maglaj is very good, and thus the quality of purified wastewater that will be discharged in river Bosna is convenient.

Concerning the influence of the input parameters or the quality of the unpurified wastewater on the value of the output parameters or the quality of the purified wastewater it can be concluded that to some extent the input parameters can affect the efficiency of the purification and the quality of the purified water. However, purification of wastewater in the cellulose and paper factory "Natron-Hayat" Maglaj does not always happen under the same conditions. If there is loaded wastewater at the entrance to the system, in order to maintain the efficiency of the purification system in the prescribed values, larger amounts of chemicals are added into the system. For this reason, it would also be necessary to monitor the amount of spent chemicals to be purified and to do the analysis.

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Pozitivni primeri efikasnosti prečišćavanja otpadnih voda u fabrici „Natron-Hayat“ Maglaj

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I Z V O D

U radu su opisane osnovne karakteristike otpadnih voda u fabrici celuloze i papira „Natron-Hayat“ Maglaj. Poseban naglasak je stavljen na opis tehnološkog procesa prečišćavanja otpadnih voda u fabrici „Natron-Hayat“ Maglaj, koji je predstavljen u radu tehnološkom šemom, a koji predstavlja jedan od složenijih sistema za prečišćavanje otpadnih voda. U eksperimentalnom delu dati su rezultati efikasnosti sistema za prečišćavanje otpadnih voda u fabrici "Natron-Hayat" Maglaj. Može se reći da su osnovne sirovine za proizvodnju papira celulozna vlakna i voda. Uloga koju voda ima u industriji celuloze i papira kao rastvarač, sredstvo za kuvanje i sredstvo za pranje dobijene celuloze, ukazuje na prisustvo vode u skoro svim fazama procesa proizvodnje celuloznih vlakana [1]. U skladu sa principima održivog razvoja, industrija celuloze i papira mora da se suoči sa strogom zakonskom regulativom o zaštiti životne sredine koja podrazumeva racionalizaciju potrošnje sirovina, vode, energije i hemikalija, uz minimalne negativne uticaje na životnu sredinu. Otpadne vode koje nastaju u fabrici „Natron-Hayat“ Maglaj podvrgavaju se prečišćavanju pre ispuštanja u površinski vodorecipijent. Otpadne vode iz fabrike „Natron-Hayat“ Maglaj se ispuštaju u reku Bosnu.