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# PULP MILL WASTEWATERS BIOLOGICAL TREATMENT\*\*

The paper presents the characteristics of pulp mill effluents. Various biological treatment methods of pulp mill wastewaters and the main advantages and disadvantages of these methods are discussed. The equations for calculation of the capital and operation costs are given. Selected examples of pilot plant experiments for kraft mill and barking effluents treatment in Finland are described.

# 1. WASTEWATERS FROM VARIOUS STAGES OF PULP MILL PROCESSES

Pulp mill produce the following kinds of wastewaters: barking effluents, washing and screening effluents, condensates, and bleaching effluents.

In the modern Finnish sulphate mills washing losses are 10 kg  $Na_2SO_4/t$ . In sulphate mills the screening system is closed, whereas in sulphite mills it is open. The bleaching system is usually CEDED or CEHDED. Wastewater loads of condensates are decreased by steam stripping in sa-mill<sup>1</sup>. In si-mills<sup>2</sup> the BOD load is reduced by yeast, protein or ethanol production. In paper mills the wastewater load depends on water circulation system, bleaching system, and on chemicals employed.

Wastewater loads from several Finnish pulp mills in the year 1976 are shown in tab. 1. The wastewater loads of modern pulp mills are given in tab. 2. The quality of pulp mill wastewaters is characterized by such parameters as BOD, COD, suspended solids, toxicity, etc. The BOD and COD of wastewaters from sa-mills are shown in tab. 3.

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<sup>2</sup> si = sulphite.

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<sup>&</sup>lt;sup>1</sup> sa = sulphate.

Wastewater loads of Finnish pulp mills in 1976 ([15]) Ładunki zanieczyszczeń w ściekach celulozowych w Finlandii w 1976 ([15])

Product		1	kg BOD/t		
Unbleached	sa-pulp		37		
Bleached	sa-pulp	· · ·	45		
Unbleached	si-pulp		92		
Bleached	si-pulp		117		
Paper			6-14		

Table 2

Wastewater loads of modern pulp mills (FLECKSEDER [6]) Ładunki zanieczyszczeń w ściekach nowoczesnych celulozowni (FLECKSEDER [6])

Product	kg BOD/t	kg COD/t
sa-pulp, conventional bleaching	25	105
sa-pulp, oxygen bleaching	17	60
sa-pulp, Rapson-Reeve	3	15
Ca-si-pulp, without burning of spent iquor	 400	1600
Ca-si-pulp, with burning of spent liquor	60-80	175-240
Mg-si-pulp, with neutralization of spent liquor	40-50	75-90

Table 3

BOD and COD of wastewaters of sa-mills ([16]) BZT i ChZT ścieków z zakładów celulozy siarczanowej ([16])

1	Wastewate	r of	mg BOD/dm <sup>3</sup>	mg COD/dm <sup>3</sup>
Unbleached	sa-pulp (settled)		400	800
Unbleached	sa-pulp (biol. treated)		67	330
Bleached	sa-pulp (settled)		260	680
Bleached	sa-pulp (biol. treated)		36	290

### 2. POLLUTION

Various components of pulp mill effluents are given in tab. 4. These components are responsible for biomass production, toxicity, increased amounts of suspended solids, higher oxygen consumption, and light absorption.

Component	Bleached sa-pulp	Bleached si-pulp	
Lignins	30	60	
Polysaccarides	3	25	
Monosaccarides		12	
Carboxyl acids	5	30	
Sugar acids and lactoses	23		
Methanol etc.	7	10	
Furtural	·	6	
Resin and fat acids	5	12	

The components of pulp mill effluents as kg/t (JÄPPINEN [10]) Składniki ścieków z zakładów celulozowych w kg/t (JÄPPINEN [10])

Biologically treated wastewaters, where BOD reduction is 70-90%, should be diluted 10-50 times (JÄPPINEN et al. [11]).

After mechanical treatment the pulp mill effluents contain 5–10 kg of suspended solids/t (in fishing waters suspended solids should be less than 25 g/m<sup>3</sup>, LANDNER et al. [14]). Lignin compounds are light absorbing. Toxic compounds are: some acids and bases, sulphides, chlorine compounds, resin and fat acids, and chlorinated lignins.

Sa-wastewaters are more toxic than si-wastewaters. Their toxicity can be reduced if instead of chlorine  $ClO_2$  or hypochlorite or oxygen bleaching is used. In biological treatment the toxicity can be reduced by about 90% (JÄPPINEN et al. [11]). Nutrients, especially nitrogen and phosphorus, increase the biomass production. Their acceptable and critical concentrations in lakes are shown in tab. 5. Table 6 presents the phosphorus loads of pulp mill wastewaters according to SÄRKKÄ ([17]). The nitrogen loads of pulp mill wastewaters according to LANDNER ([13]) are shown in tab. 7.

#### Table 5

#### The acceptable and critical concentrations of N and P (g/m<sup>2</sup> a) in lakes depending on the depth (m) of the lake (VOLLENWEIDER [18])

Dopuszczalne i krytyczne stężenia N i  $P(g/m^2 a)$  w jeziorach

w zależności od głębokości (m) (Vollenweider [18])

Depth		Acceptable			C	Critical	
	m	Ν	Р	i.	Ν	Р	
	5	1.0	0.07		2.0	0.13	
	10	1.5	0.10		3.0	0.20	
	50	4.0	0.25		8.0	0.50	

Phosphorus loads of pulp mill wastewaters (Särккä [17]) Ładunki fosforu w ściekach z celulozowni (Särккä [17])

Process		g P/t
Unbleached	si-pulp	100
Bleached	si-pulp	100
Bleached	sa-pulp	115

#### Table 7

Nitrogen loads of pulp mill wastewaters (LANDNER [13]) Ładunki azotu w ściekach z celulozowni (LANDNER [13])

Process	g N/t
Unbleached sa-pulp	260-530
Unbleached si-pulp	280-620

# 3. BIOLOGICAL TREATMENT

Biological treatment of pulp mill wastewaters can be divided in the following way: aerated lagoon, extended aeration, conventional activated sludge process, biofilter, and anaerobic treatment.

#### 3.1. AERATED LAGOON

In aerated lagoon the hydraulic residence time is 5-20 days and the water depth 3-5 m. The wastewaters are aerated by mechanical aerators or by compressed air. The tasks of aerators are: to supply oxygen and to keep suspended solids in mixture.

The pressure aeration can be: coarse bubble aeration, medium bubble aeration, and fine bubble aeration. Mechanical aerators are either rapidly or slowly rotating, the slowly rotating mechanical aerators being the most economical are used more often. While designing aerated the lagoon the following factors should be considered: BOD removal, temperature, oxygen demand, excess sludge, nutrient demand, and mixing.

Pulp mill wastewaters biological treatment

The BOD removal in aerated lagoon is shown in fig. 1. According to BODENHEIMER [2] the effluent from an aerated lagoon can be calculated from the following equation:

$$BOD_0 = [(467 - 0.8027 \ T)/T]^d BOD_i$$
, (1)

where:

 $BOD_0$  — effluent BOD (mg/dm<sup>3</sup>), BOD<sub>i</sub> — influent BOD (mg/dm<sup>3</sup>), T — temperature in lagoon (K), d — hydraulic residence time (d).



Fig. 1. BOD removal in aerated lagoons (CARPENTER et al. [3]) Rys. 1. Usuwanie BZT w lagunach napowietrzanych

(CARPENTER et al. [3])

The temperature coefficient  $\Theta$  in aerated lagoon is 1.06–1.09, whereas in activated sludge plants it varies within 1.00–1.03 (JANK [8]). The BOD removal in aerated lagoon is a function of volume load (fig. 2), and the oxygen consumption is 0.6–1.7 kg O<sub>2</sub>/kg BOD remo-



Fig. 2. BOD removal in aerated lagoon as a function of volume load (JÄPPINEN et al. [11]) Rys. 2. Usuwanie BZT w lagunach napowietrzanych w funkcji obciążenia objętościowego (JÄPPINEN et al.) [11]

ved. Oxygenation capacity according to EMDE [5] is shown in tab. 8. The nozzles often used are Kenics and Helixor and in Finland Nokia. The common slowly rotating aerators are Simcar, Vortair, Lightnin, Gyrox, Simplex, BSK, and Actirotor. Their oxygenation capacity for tap water is  $1.5-2.0 \text{ kg O}_2/\text{kWh}$ .

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# Oxygenation capacity (g $O_2/m^3$ m) of compressed air [EMDE [5])

Pojemność nasycenia tlenem sprężonego powietrza  $(g O_2/m^3 m)$  (EMDE [5])

Aeration system	g	$O_2/m^3$	m
Fine bubble aeration		7	1
Medium bubble aeration		4.5	
Coarse bubble aeration		4	

The amount of excess sludge without endogenous respiration is 0.5-0.8 kg/kg BOD removed, and 20-70% of sludge is discharged in effluent of aerated lagoon.

The demand of nutrients in aerated lagoons is the smallest one if compared with other biological processes in which the normally applied ratio of BOD:N:P = 100:5:1. In aerated lagoons the nutrients can be re-used.

The purpose of aeration is also to keep the mixed liquor in suspension. According to WIGHT [19] the adequate mixing energy is  $3 \text{ W/m}^3$ , if the amount of suspended solids is 500 mg/dm<sup>3</sup>. According to BEAK [1] the demand of mixing energy is  $2.8-11.8 \text{ W/m}^3$ . The bottom scour velocity should be more than 0.12-0.15 m/s (ECKENFELDER [4]).

The aerated lagoons have the following advantages: they are suitable to every type of pulp mill effluents, easy to operate, the capital costs are relatively low and nutrient demand is small.



Fig. 3. The capital costs of aerated lagoons (BOD removal = 75%) and activated sludge plants (BOD removal = 90%) (JÄPPINEN et al. [11])

Rys. 3. Nakłady inwestycyjne na laguny napowietrzane (usuwanie BZT = 75%) i oczyszczanie metodą osadu czynnego (usuwanie BZT = 90%) (JÄPPINEN et al. [11])

The main disadvantages of aerated lagoon are: a large area needed, the presence of suspended solids in effluent if no secondary settling tank is used (badly settleable solids), and lower BOD removal than in activated sludge process.

JÄPPINEN et al. [11] have calculated the capital and operation costs of aerated lagoons from the equations (2) and (3):

$$K = 0.5 \, Q^{0.73} S_r^{0.3} \left(\frac{S_r}{S_i}\right)^3,\tag{2}$$

$$k = 0.0088 \ Q^{0.85} \mathrm{S}_r^{0.7} \; ,$$

where:

K - capital costs (1000 dollars),

O – amount of wastewater (m<sup>3</sup>/d),

 $S_{\star}$  – BOD removed in 25°C (mg/dm<sup>3</sup>),

 $S_i$  – BOD of influent (mg/dm<sup>3</sup>),

k - operation costs (1000 dollars/a).

Figures 3 and 4 show the capital and operation costs of aerated lagoons. The cost of sludge pumping from the lagoon is included, while the design cost and cost of wastewater transfer to the lagoon and that of sludge dewatering (40–50 dollars/t) are not included. The construction of the lagoon amounting to 5–10 dollars/m<sup>3</sup> of tank is the main capital cost (50-70%).



Fig. 4. The operation costs of aerated lagoons (BOD removal = 75%) and activated sludge plants (BOD removal = 90%) (JÄPPINEN et al. [11])

Rys. 4. Koszty eksploatacyjne dla lagun napowietrzanych (usuwanie BZT = 75%) i oczyszczalni pracujących metodą osadu czynnego (usuwanie BZT = 90%) (JÄPPINEN et al. [11])

(3)

#### 3.2. ACTIVATED SLUDGE PROCESS

The types of activated sludge process depending on the loading are shown in tab. 9. The activated sludge plants for pulp mill wastewaters are normally designed for a sludge load of 0.2–0.5 kg BOD/kg MLSS d and for MLSS 1.5–4 kg/m<sup>3</sup>. Overflow rate in secondary settling tank ranges between 0.6–1.2 m/h. According to EMDE [5] oxygenation capacity can be calculated by using the following equation:

$$OC = \alpha BOD,$$

where:

OC – oxygenation capacity (kg  $O_2/d$ ), BOD – BOD load (kg BOD/d), a – safety coefficient.

#### Table 9

(4)

Types of activated sludge process (EMDE [5]) Rodzaje procesu osadu czynnego (EMDE [5])

Process	$T^1$	$L_{\nu}^{2}$	MLSS <sup>3</sup>	$L_s^4$	BOD removal, %
High rate	1.5	3.6	3.6	1.0	80
Conventional	6	0.7	2	0.35	90
Extended aeration	24	0.36	3.6	0.1	90

 $^{1}T$  – aeration time (h).

 $^{2}L_{y}$  - volume load (kg BOD/m<sup>3</sup> d).

<sup>3</sup>MLSS – mixed liquor supsended solids (kg/m<sup>3</sup>).

 $^{4}L_{s}$  – sludge load (kg BOD/kg MLSS d).

The safety coefficient  $\alpha$  for high rate process, conventional process, and extended aeration amounts to 1.2, 1.5, and 2, respectively. In the design of activated sludge process for pulp mill wastewaters the following parameters must be taken into account: temperature, pH, toxic substances, and nutrients.

The temperature should be lower than  $35^{\circ}$ C. The activated sludge plants operate also well at low temperatures (2–5°C). The best pH is within 6.5–8.5. Toxic substances have been discussed earlier. The toxicity of pulp mill effluents does not normally inhibit biological treatment. The nutrients are needed in a ratio of BOD:N:P = 100:5:1. If the sludge load is low, the nutrient demand is also smaller. The amount of excess sludge is 0.4–0.7 kg/kg BOD removed. Water content in excess sludge is high - 98.5–99.5%. The excess sludge

must be thickened and dewatered mechanically.

The activated sludge process for pulp mill effluents has the following disadvantages: Lignin compounds cannot be removed.

Nutrients that must be dosed cause tertiary pollution.

Capital and operation costs are high.

Amount of the excess sludge is high.

BOD removal can be reduced by fluctuations in wastewater quantity and quality.

Capital and operation costs are shown in tabs. 10 and 11. JÄPPINEN et al. [11] have calculated the capital and operation costs of activated sludge plants from the equations (5) and (6).

Table 10

Koszty inwestycyjne (%) (JÄPPINEN et al. [11])					
	Activated sludge	Biofilter	Aerated lagoon		
Construction	40-55	30-40	50-70		
Machinery	30-40	50-65	20-40		
Electricity etc.	10-20	8–10	8 -10		

Capital costs (%) (JÄPPINEN et al. [11])

Table 11

Operation costs (%) (JÄPPINEN et al. [11]) Koszty eksploatacji (%) (JÄPPINEN et al. [11])

	Activated	Biofilter	Aerated lagoon	
Labour	25	30	15	
Energy	25	15	50	
Nutrients	20	25	10	
Other chemicals	5	5	—	
Maintenance	10	15	10	
Administration	10	10	15	

$$K = 0.13 \ Q^{0.75} \ S_r^{0.45} \ rac{S_r}{S_i} \ ,$$

$$k = 0.00085 Q^{0.85} S_r^{0.8}$$

(5) (6)

where:

K - capital costs (1000 dollars),

Q – amount of wastewater (m<sup>3</sup>/d),

 $S_r - BOD$  removed (mg/dm<sup>3</sup>),

 $S_i$  – BOD of influent (mg/dm<sup>3</sup>),

k – operation costs (1000 dollars/a).

Figures 4 and 5 show the capital and operation costs of activated sludge plants. These figures do not include design costs and costs of leading of wastewaters to the plant.

#### 3.3 BIOFILTER

Biofilter consists of 3–10 m high tower filled with plastic particles or stones. Wastewater is usually 1–4 times recycled through the filter. After the filter there is a secondary settling tank. Primary settling tank is necessary, because otherwise the filter pipes will be blocked. The volume load is 2–5 kg BOD/m<sup>3</sup>d and hydraulic load 20–100 m<sup>3</sup>/m<sup>3</sup>d. BOD removal is 50–80%. Nutrient demand and the amount of excess sludge are the same as in the activated sludge process. Biofilters are well resistant to toxicity. The disadvantage of biofilter is that the BOD removal is relatively low.

JÄPPINEN et al. [11] have calculated the capital and operation costs of biofilters from the equations (7) and (8):

$$K = 0.036 \ \underline{Q}^{0.8} S_i^{0.6} L_{\nu}^{-0.5} , \qquad (7)$$
  
$$k = 0.00055 (OS_{\nu})^{0.85} , \qquad (8)$$

where:

K — capital costs (1000 dollars),

k - operation costs (1000 dollars/a),

Q – amount of wastewater (m<sup>3</sup>/d),

 $S_i$  – BOD of influent (mg/dm<sup>3</sup>),

 $S_r$  – BOD removed (mg/dm<sup>3</sup>),

 $L_v$  – volume load (kg BOD/m<sup>3</sup>d).

#### 3.4. ANAEROBIC TREATMENT

In anaerobic treatment the substrate is first decomposed to alifatic acids (e.g. acetic acid) and then to  $CO_2$  and  $CH_4$  by methane bacteria. The optimum temperature is 35°C. Anaerobic treatment is economical, if influent BOD is higher than 1000–2000 mg/dm<sup>3</sup>. Few pulp mill effluents have so high a concentration, only condensates of si-mills and barking effluents in a closed system. Anaerobic treatment can take place in a reactor or in an anaerobic filter.

The advantages of anaerobic treatment are: low energy consumption, production of methane, small amount of excess sludge, and low nutrient demand.

Anaerobic treatment have also some disadvantages:

it requires higher substrate concentration,

it is sensitive to fluctuations in wastewater quality.



JÄPPINEN et al. [11] calculated the capital and operation costs of anaerobic treatment from the equations (9) and (10):

$$K = 0.0062(QS_i)^{0.85}, (9)$$
  

$$k = 0.0002(QS_i)^{0.85}. (10)$$

where:

K — capital cost (1000 dollars),

k — operation cost (1000 dollars/a),

Q – amount of wastewater (m<sup>3</sup>/d),

 $S_i$  — influent BOD (mg/dm<sup>3</sup>).

# 4. PILOT PLANT EXPERIMENTS IN FINLAND

# 4.1. EXTENDED AERATION PILOT PLANT WITH PERIODICALLY OPERATING SECONDARY SETTLING TANKS FOR KRAFT MILL EFFLUENTS

Unbleached and bleached kraft mill effluents from two factories were treated (HIDEN-HEIMO [7]) in a 300 m<sup>3</sup> pilot plant with two periodically operating settling tanks and brush aeration (fig. 5). Volume load in the Lohja experiments was 0.2–0.3 kg BOD/m<sup>3</sup> d, sludge load 0.07–0.17 kg BOD/kg MLSS d, MLSS 1.7–3.4 kg/m<sup>3</sup>, aeration time 10–14 h, and overflow rate 0.8–1.2 m/h. The Lohja mill produced unbleached kraft pulp. Wastewater was not pretreated and its BOD was 100–170 mg/dm<sup>3</sup> and pH neutral. The BOD removal



Fig. 5. The extended aeration pilot plant used in Lohja and Kaukas, the scale in centimeters (HIIDENHEIMO) [7])

Rys. 5. Eksperymentalna oczyszczalnia ścieków pracująca metodą osadu czynnego z przedłużonym napowietrzaniem zastosowana w Lohja i Kaukas, skala w centymetrach (НПДЕNНЕІМО [7]) of 80–95% could be achieved by dosing phosphorus in a ratio of BOD: P = 100:0.3. In the Kaukas experiments wastewater of bleached kraft process was treated. Volume load was 0.2 kg BOD/m<sup>3</sup> d, sludge load 0.09 kg BOD/kg MLSS d, MLSS 2.5 kg/m<sup>3</sup>, aeration time 14 h, and overflow rate 0.8 m/h. Although no nutrients were dosed and pH was not adjusted, the BOD removal was as high as 70%.

#### 4.2. EXTENDED AERATION PILOT PLANT FOR BARKING EFFLUENT

KALLIOLA [12] treated the effluents of two barking plants. The Voikkaa plant (fig. 6) had an open water system (4.3–6.5  $m^3/m^3$  of timber), and the Myllykoski plant a closed water system (1.0–1.2  $m^3/m^3$  of timber). BOD of wastewater was 500–3000 mg/dm<sup>3</sup> and MLSS 2–17 kg/m<sup>3</sup>. The volume of the pilot plant was 2.5 m<sup>3</sup>. The following observations have been made:



Fig. 6. The pilot plant of barking effluents (KALLIOLA [12]) Rys. 6. Eksperymentalna oczyszczalnia dla ścieków z odkorowialni (KALLIOLA (12])

pH of wastewater greater than 4.0 need not be regulated.

In sludge load greater than 0.5 kg BOD/kg MLSS d, the BOD removal decreases rapidly. To achieve 85-95%, BOD removal of the sludge load must be less than 0.05 kg BOD/kg MLSS d.

Sludge is badly settleable and needs large secondary settling tank.

Nutrient demand is BOD: N:P = 100:2:0.3. Barking effluent contains enough phosphorus but not always enough nitrogen.

The use of lime or Al-sulphate in aeration tank decreases BOD removal.

# 4.3. PILOT PLANT FOR ALKALINE BLEACHING EFFLUENTS

JUNNA et al. [9] treated alkaline bleaching effluents in a pilot plant with an 11 m<sup>3</sup> aeration tank. The average BOD of wastewater was 600 mg/dm<sup>3</sup>. Volume load varied within 0.5–4.5 kg BOD/m<sup>3</sup> d. Good results could be obtained if the volume load was less than 2 kg BOD/m<sup>3</sup> d and BOD:N:P = 100:2:0.5. Due to high fluctuations in wastewater quality the operation with the pilot plant was difficult. Thus, it is better to treat more than one wastewater fraction.

# 5. ADVANTAGES AND DISADVANTAGES OF BIOLOGICAL TREATMENT

In 1980 there were in Sweden 6 aerated lagoons, 2 biofilters, and 2 activated sludge plants, whereas Finland had 6 aerated lagoons. Finnish pulp and paper mills consumed 3 500 000 m<sup>3</sup> of water/d, and the BOD load was 1000 t/d. JÄPPINEN et al. [11] have calculated that costs (capital and operation) of aerated laggoon in sa-mill (200 000 t/a) are 6–9 dollars/t of pulp, in si-mill (100 000 t/a) 9–11 dollars/t of pulp and the cost of activated sludge plant in paper mill (150 000 t/a) are 5–7 dollars/t of paper.

The performance of the plants has shown that biological treatment is characterized by removal of toxicity and rapidly oxygen consuming compounds.

The disadvantages of biological treatment are the following:

It does not remove colour and lignin compounds.

It may cause a tertiary pollution due to the nutrients.

It is sensitive to fluctuations in wastewater quality.

It is rather expensive.

There are the following treatment methods: Al-sulphate precipitation, lime precipitation, active carbon adsorption, ion-exchange, and ultrafiltration. These methods are expensive and therefore not commonly used.

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# BIOLOGICZNE OCZYSZCZANIE ŚCIEKÓW CELULOZOWYCH

Scharakteryzowano ścieki z celulozowni. Omówiono zastosowanie wielu metod biologicznego oczyszczania tych ścieków. Podano zarówno zalety i wady poszczególnych metod, jak również równania pozwalające na obliczenie nakładów inwestycyjnych i kosztów eksploatacyjnych dla poszczególnych procesów oczyszczania. Opisano wybrane przykłady badań eksperymentalnych w oczyszczaniu ścieków z zakładów celulozowych na stacjach pilotowych w Finlandii.

# BIOLOGISCHE KLÄRUNG VON ZELLSTOFFABWÄSSERN

Im Aufsatz wurde das Zellstoffabwasserproblem, vor allem die Anwendung verschiedener biologischer Methoden der Abwasserklärung besprochen. Sowohl Vorzüge wie auch Nachteile einzelner Methoden wurden hervorgehoben und Gleichungen angegeben, die die Berechnung von Investitions- und Betriebskosten einzelner Klärungsprozesse ermöglichen. Ausgewählte Beispiele experimenteller Untersuchungen über Zellstoffabwasserklärung, die in finnischen Untersuchungsstationen durchgeführt worden waren, wurden beschrieben.

# БИОЛОГИЧЕСКАЯ ОЧИСТКА СТОЧНЫХ ВОД ПРЕДПРИЯТИЙ / ЦЕЛЛЮЛОЗНОЙ ПРОМЫШЛЕННОСТИ

В работе охарактеризованы сточные воды, сбрасываемые предприятиями целлюлозной промышленности. Рассматривается применение ряда методов биологической очистки этих вод. Приводятся преимущества и недостатки отдельных методов, а также уравнения, позволяющие вычислить капиталовложения и эксплуатационные затраты для отдельных процессов очистки. Описаны избранные примеры экспериментальных исследований на пилотажных станциях в Финляндии для очистки сточных вод предприятий целлюлозной промышленности.