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DISINFECTION BYPRODUCTS PRECURSORS REMOVAL FROM DAM RESERVOIR WATER

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Abstract: The water of the Wisła-Czarne reservoir is of very low hardness and alkalinity. In spite of high SUVA values it is not susceptible to enhanced coagulation. In order to achieve the assumed results, coagulation with ALS (aluminium sulphate) should be conducted in two optional technological systems – in a conventional system and in "in-bed" coagulation dependently of water quality and its temperature. Effective treatment with ALS is possible, even at low temperature of the water, but at strict technological parameters. However, because of significant variations of water quality, especially after rainstorms, it is very difficult to meet such requirements. Application of pre-hydrolyzed Flokor 1,2A instead of ALS enables to eliminate reagents to the pH adjustment and to apply "in-bed" coagulation when water supplied to the WTP is of low turbidity. To assure stable technological system operation, in aspect of raw water quality changes, some activities were also undertaken, i.e. modernization of rapid filters, which involved a drainage system and exchange of sand bed for anthracite-sand bed. Treatment based on direct filtration results in decrease of reagents usage and, what is especially important, effective DBPs precursors removal.

INTRODUCTION

Organic matter in natural waters contains both hydrophobic and hydrophilic fractions of NOM. The hydrophobic fractions are generally composed of higher molecular weight NOM with activated aromatic rings, phenolic hydroxyl groups and conjugated double bonds, while the hydrophilic fractions are typically composed of the lower molecular weight NOM with aliphatic ketones and alcohols [9, 20]. The hydrophobic fractions of NOM exhibit higher ultraviolet absorbance (UV_{254}) and higher SUVA (Specific UV Absorbance) while the hydrophilic fractions of NOM exhibit lower UV_{254} and lower SUVA. The SUVA is an operational indicator which, on the one hand allows to determine the characteristics of NOM, and on the other hand, the effectiveness of coagulation in removal of NOM, TOC and DBPs (Disinfection By-Products) precursors [12]. For waters with low SUVA ≤ 2.0 $dm^3/mgC \cdot m$ organics are mainly of non-humic nature and they are not susceptible to "enhanced" coagulation.

Pretreatments prior to chlorination can partially remove NOM and this removal can be enhanced by using GAC or enhanced coagulation. Introducing alternative disinfectants or a combination of disinfectants (chloramine, ozone, chlorine dioxide and ultraviolet radiation followed by post chlorination) can also reduce formation of DBPs. However, the use of these alternative disinfectants can still lead to the formation of more toxic DBPs. More than 118 models to predict DBPs including trihalomethanes (THMs), haloacetic acids (HAAs), haloacetonitriles (HANs), and haloketones (HKs) formations in drinking waters have been reported. The parameters frequently incorporated in the development of DBP predictive models include total organic carbon (TOC), dissolved organic carbon (DOC), ultraviolet absorbance at 254 nm (UV_{254}), specific ultraviolet absorbance (SUVA), pH, temperature, bromide ion concentrations, chlorine dose and reaction time. It was stated that the longer reaction time (the higher consumption of residual disinfectant) the more DBPs are produced. However, some research indicate that chlorinated DBPs such as HAAs may degrade in endings of distribution systems [6, 25, 26]. The effect of the pH is variable for different by-products. THM concentration increases with the increase of the pH. For HAA the effect is opposite. The higher pH the lower concentration of HAA is observed [28]. It has been also proved that higher temperature increases the rate of DBP formation. Singer and Chang [27] stated linear relationship between TOX (Total Organic Halogens), THM, UV_{254} and TOC. There have been many other investigations which developed the relationships between precursors and operational indicators and DBPs [1, 6, 14, 15, 28]. Korshin, Li and Benjamin [19] stated that there is a linear correlation between the decrease of absorbance UV at 272 nm and TOX formation. This relationship was confirmed for waters of variable quality with wide range of DOC (dissolved organic carbon), at various chlorine doses, various duration of chlorination and at wide range of pH = 5–11. The equation is as follows:

$$\mathrm{TOX} = 10834\ \Delta UV_{272} \tag{1}$$

Enhanced coagulation is an effective method for DBPs precursors removal, especially for high MW organic compounds (> 30 kDa) [11, 18, 24, 32]. Aluminium chloride ($AlCl_3$) and polyaluminum chloride (PACl) are common used coagulants for DBPs precursors removal. Many researchers believe that PACl is more effective in removing turbidity, dissolved organic carbon (DOC), and UV_{254} absorbance than traditional aluminium coagulants. PACl is characterized by high positive charge and strong binding ability. Preformed Keggin-Al_{13} compounds are regarded to be the most active species responsible for coagulation. Some research proved that traditional aluminium salts ($AlCl_3$) generated this type of aluminium polymeric species at pH 5.0–6.0 in situ. $AlCl_3$ with a high content of in situ transformed polymeric species was more effective than PACl in removing DOC and UV_{254} [5, 6, 7, 29].

It was also found that a combination of flocculant and coagulant enhanced the coagulation-flocculation process and humic acid removal. The optimum conditions of coagulation-flocculation were established in reference to the ratio of humic acid and coagulant. The ratio of E4/E6 (the ratio of absorbance at 465 nm and 665 nm) shows the molecular size variations using different coagulants and flocculants [8, 33].

CHEMICAL CHARACTERISTICS OF THE BIAŁA AND THE CZARNA WISEŁKA

The literature data present classification of natural waters on the basis of mineralization, concentration of total solids and dissolved substances. Waters of low mineralization consist of up to 3 g/dm^3 of total solids. The maximal concentration of dissolved substances in these waters is 200 mg/dm^3. Such waters are met in mountainous areas. In the years 1993–1995 the Institute of Nature of the Polish Academy of Sciences in Cracow made the chemical research of both Wisełka rivers. The results indicate that the waters of the Czarna Wisełka are acidified, of low pH, sometimes they lack alkalinity and they are also characterized by very low concentration of calcium. High concentrations of aluminium were also noted there. The highest acidification, the highest aluminium and organic matter concentration were observed in the period of snow melting and after rainstorms. With the course of the Czarna Wisełka its chemical characteristic changed. Concentration of calcium and magnesium acid carbonate increased, so did pH, but aluminium concentration decreased. The Biała Wisełka waters distinctly differed from the Czarna Wisełka ones. The pH was over 6.0, alkalinity always exceeded 0.25 mval/dm^3. Taking into consideration the basin of both Wisełkas, ionic composition of water in the reservoir was influenced by the waters of the Biała Wisełka.

The reservoir drainage area mainly consists (88%) of forests. The soil testing showed that the Czarna Wisełka basin is covered mainly by podsol produced on poor calcium and magnesium sandstone of the Istebnian stratum. In the Biała Wisełka basin there is more brown soil produced from sandstones and mudstones. The Czarna Wisełka basin soil is more acidic and of higher exchangeable aluminium concentration. The Biała Wisełka basin soil reveals higher concentration of exchangeable cations, so a higher calcium concentration in water flowing from this area is noted [21, 22]. Last century spruce monoculture replaced mixed stand resulting in acceleration of soil podzolization [10]. The intensive rainfalls were also the additional agent which enhanced that process. The rainfalls washed the shallow soil and caused soil leaching from alkalies [16]. It was the reason of higher soil sensitivity to inflow of acidic compounds originated from the atmosphere contamination.

In 1981–1983 Pająk [23] conducted long-term investigations on both net and sedimented phytoplankton samples collected in the Wisła-Czarne Dam Reservoir and its bays at the Biała Wisełka and at the Czarna Wisełka (Western Carpathians). Wróbel [31] emphasized that the occurrence of "water bloom" above the reservoir was observed periodically with the inflow of acidified water (mainly after snow melting and heavy rainfall). It suggested that acidification did not prevent eutrophication. "Water bloom" in the reservoir was associated with increased acidification, which caused a decrease in biodiversity [2, 3, 4, 23].

WATER CHARACTERISTICS IN THE WISŁA-CZARNE RESERVOIR

The Wisła-Czarne dam reservoir was built on the Vistula River in the area where the Biała and the Czarna Wisełka connect. It is the highest located dam reservoir in Poland [30]. The reservoir plays a significant role in supplying water to some cities of great recreation importance. The dam of the maximal height of 36 m and the length of 280 m is situated about 300 m beneath the place where both Wisełka rivers meet. The basic role of the reservoir is compensation of strongly changing water flow. The total capacity of reservoir is 5.06 mln m^3, of maximal area 40 ha. The retention time of the Wisła-Czarne reservoir at the typical water level (544.4 m npm) and corresponding capacity 2.1 mln m^3 and average flow of the both rivers 0.825 m^3/s is 29 days. Water is taken from three levels dependently of its quality. Water recirculation in the reservoir strongly depends on the bottom outlet. In 1995 the reservoir was emptied because of the repair work of its construction. The work lasted for some years until the beginning of 1999. Since then for some months the level of water in the reservoir had been changing till autumn 1999 [17]. Temperature measurements and dissolved oxygen concentration at the close-to-dam part of the reservoir show the lack of typical for lakes thermal stratification. When the reservoir was under modernization the pH of the water was much higher (pH = 6.9–7.1) than in the following year after its filling (pH = 5.5–6.3). In 2001 pH significantly increased and was changing in the range of 6.5–7.35. Such pH variations probably result from the fact that the reservoir started its new operation as a biological system. In the first period after the filling, the reservoir was a typical oligosaprobic reservoir and some time was needed to increase its fertility. At that time the compounds, that leached from the bottom and from the reservoir construction during its filling, strongly influenced the pH values. The results indicate that these compounds must have had alkaline properties. After completing the process of the reservoir filling, when the amount of water in the reservoir was stable, the pH value decreased, because it was only dependent on the quality of the Wisełka waters. Later phytoplankton developed, microorganisms became more and more significant in carbon dioxide circulation. Because of low buffer capacity of the water, the changes of carbon dioxide concentration were of great importance for the pH values. Therefore, it could be assumed that in the reservoir there was a group of microorganisms which was able to absorb CO_2 and stabilize its concentration at the level lower than it resulted from that gas concentration in the water supplying the reservoir.

The water of the reservoir is qualified as low mineralized. Its conductivity is in the range of 45–90 μS/cm. Maximal hardness is 60 mg $CaCO_3/dm^3$, alkalinity is noted at very low level 0.4–0.7 mval/dm^3. It means that the water is of low buffer capacity, which is especially important in conventional coagulation with hydrolyzing coagulants, because it is very difficult to maintain proper technological parameters of a treatment process. In the analyzed research period the highest POD (Permanganate Oxygen Demand) value was 7.4 mg O_2/dm^3 with corresponding turbidity 16.8 NTU and colour 55 mg/dm^3. The highest turbidity (120 NTU) in the water of the reservoir after its filling was noted in August 2006. Very high values of turbidity were incidentally noted after rainstorms. However, the average values did not exceed 6 NTU. The highest colour was 82 mg/dm^3 and average annual values were 30 mg/dm^3. Figure 1 presents the characteristics of the water in the reservoir.

The analysis of UV_{254} absorbance in filtered (0.45 μm) and unfiltered samples, measurements of TOC (Total Organic Carbon), DOC (Dissolved Organic Carbon) showed that from the moment of the reservoir filling organic matter in water was mainly in a dissolved form. The highest amount of THMs precursors in raw water collected from the reservoir was measured in autumn. Chloroform concentration after 30-min-chlorination was 113 μg/dm^3, after 24 hour-chlorination time – 210 μg/dm^3. The lowest precursors concentration was in a winter-spring season. In summer time maximal chloroform concentration was 75 μg/dm^3. Chlorine doses applied in the testing were the same as the disinfectant

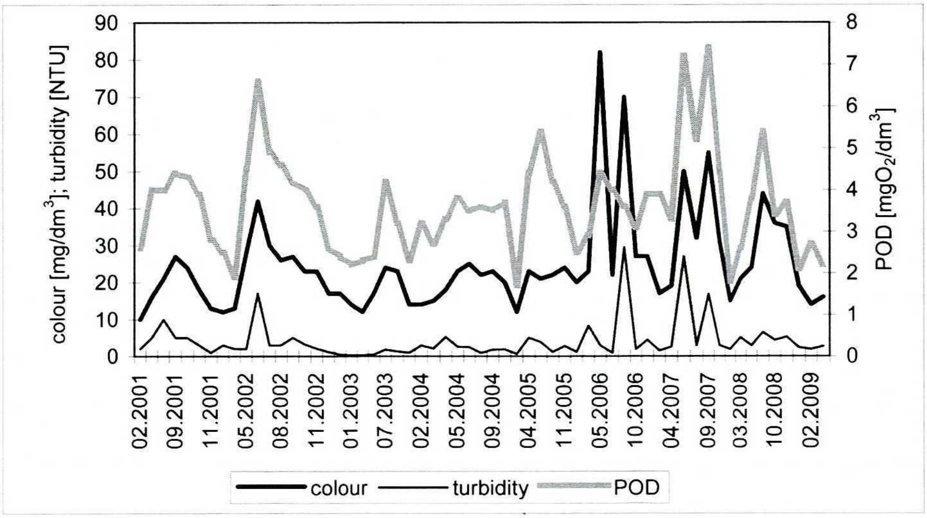

Fig. 1. Water quality in the Wisła-Czarne reservoir (after its filling) in the years 2001–2009

doses used at the Wisła-Czarne WTP (Water Treatment Plant). The results showed that 80% of chloroform was produced within the first 2 hours, the process was completed within 6 hours. Hence, it might be concluded that in water the distribution system, when water is transported for long distances, the amount of THMs should not distinctly increase. However, no correlation was found between TOC and chloroform concentration (Fig. 2).

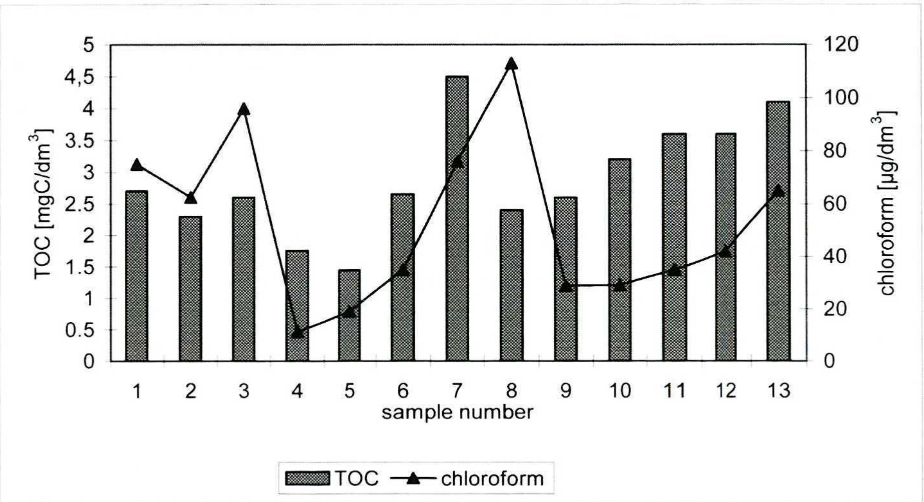

Fig. 2. TOC and chloroform concentration (after chlorination) in waters of the Wisła-Czarne reservoir

SUVA values were noted in the range of 2.0–4.9 dm^3/mg C·m which confirmed that the water was rich in DBPs precursors. It also indicated the water susceptibility to "enhanced" coagulation. The research proved that in waters where SUVA was higher than 3.0

dm^3/mg C·m humic fractions were predominant compounds, because they easier reacted with chlorine than fulvic acids [13]. Fig. 3 presents changes of chloroform concentration in the water of the Wisła-Czarne reservoir vs SUVA.

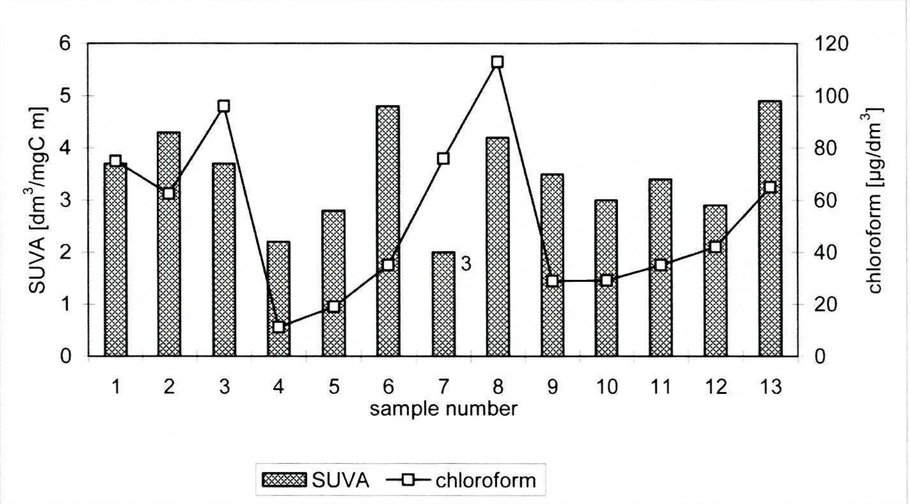

Fig. 3. SUVA and chloroform concentration (after chlorination) in the water of the Wisła-Czarne reservoir

THE ANALYSIS OF OPERATIONAL PROBLEMS AT THE WISŁA-CZARNE WTP

The treatment system at the Wisła-Czarne WTP was designed as a conventional treatment system (a hydraulic tank of rapid-mixing, vertical sedimentation tanks combined with rotational flocculators and rapid filters). Disinfection is run with the usage of low-pressure UV lamps and sodium hypochloride (NaOCl). Taking into consideration high aggressiveness of treated water corrosion inhibitors are also applied. Inhibitors are added to minimize the risk of secondary contamination by corrosion products. Requirements for coagulation effectiveness in the analyzed technological system are very high, because of high values of THMFP (Trihalomethans Formation Potential) and lack of ozonation and active carbon filtration. So that to estimate treatment effectiveness the research in two technological systems was made (conventional treatment and direct filtration). The system of conventional coagulation consisted of a rotational flocculation chamber, a vertical sedimentation tank and a pressure rapid filter. The system of direct filtration involved a pressure rapid filter. The dual media filter were applied in both systems (sand – 0.8–1.2 mm and anthracite – 0.6–2.0 mm). Each layer was 55 cm high. Both systems were supplied with the source water directly from the water intake. Operating parameters of the system tested in a pilot scale are presented in Table 1.

The water quality was evaluated on the basis of the pH, colour, turbidity, POD (Permanganate Oxygen Demand) and UV$_{254}$ absorbance and chloroform concentration. The results of the research showed that the effectiveness of coagulation depended mainly on a proper choice of technological parameters of the process [17]. Taking into consideration

Table 1. Operational characteristics of technological systems tested in a pilot scale

System	Parameter	Range of tested values	Optimum value
Conventional treatment			
Flocculator	retention time [min] flow velocity [mm/s]	12.2–20 1.7–2.8	18.3 1.9
Sedimentation tank	retention time [min] ascending velocity in the clarification zone [mm/s]	106–175 0.2–0.33	160 0.22
Filter	filtration rate [m/h]	5–8	6
Direct filtration			
Filter	filtration rate [m/h]	3–12	6

the variations of raw water quality, the optimum dose was stated just before the beginning of each testing series. Because of low mineralization and lack of buffer capacity of treated water, a very precise pH adjustment was required during a hydrolyzing coagulant (aluminium sulphate) dosing. It was stated that the optimum pH for conventional treatment was 6.0–6.5. When only ALS was applied (at the effective dose) pH dropped from 6.5 to 3.5. The optimum pH for direct filtration was in the narrow range of 6.0–6.2. Figures 4 and 5 present the effect of pH on residual aluminium concentration after treatment in the conventional system and in "in-bed" coagulation.

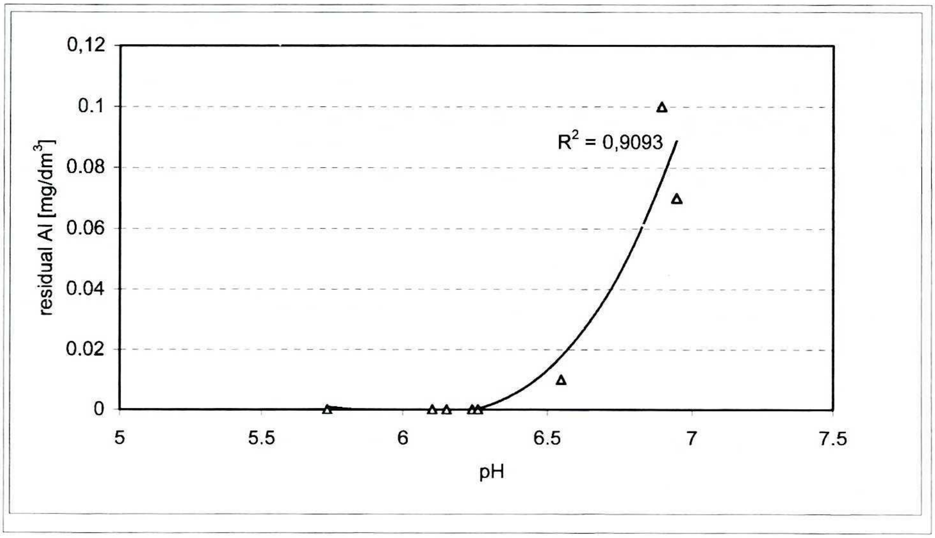

Fig. 4. Effect of pH on residual aluminium after conventional treatment

To optimize the technological process it was necessary to use both hydrolyzing coagulant and alkalies or to change the type of coagulant. Characteristics of water supplied to the distribution system at the Wisła-Czarne WTP confirm that at the optimum technological parameters of coagulation, the removal of THMs precursors was effective (Fig. 6). The acceptable level of THMs for drinking water was exceeded only in the first year after the reservoir modernization had been finished. In the later period maximal chloroform

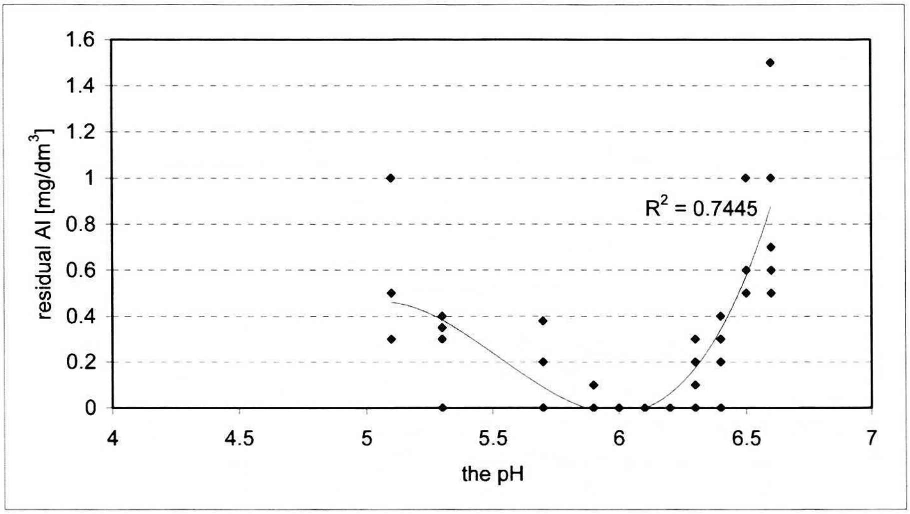

Fig. 5. Effect of pH on residual aluminium after treatment in „in-bed coagulation"

concentration in water supplied to the distribution system was 15 μg/dm^3. However, no correlation between color or POD values and chloroform concentration could be stated (Figs 7 and 8).

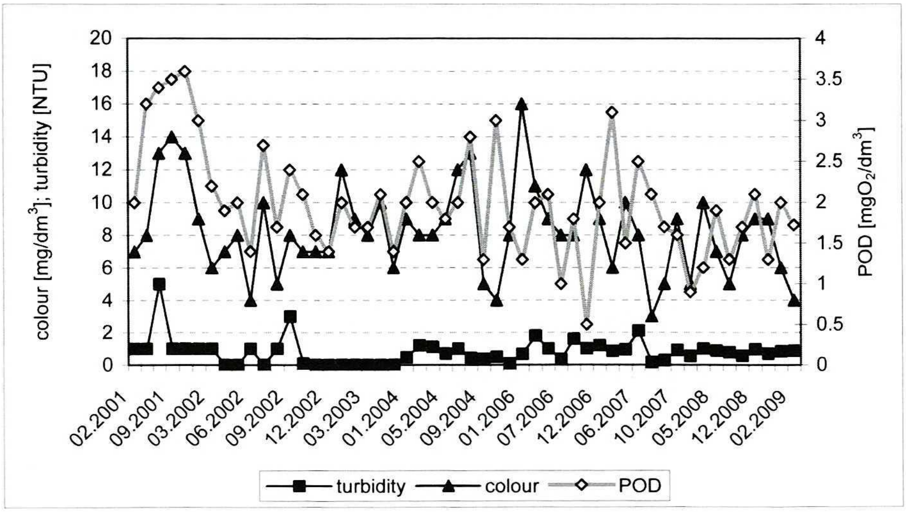

Fig. 6. Characteristics of the water supplied to the distribution system at the Wisła-Czarne WTP

Because of significant variation of water quality, especially during rainstorms, treatment with aluminium sulphate caused a lot of technological problems. The effective coagulant dose was stated in a very narrow range, so it was extremely difficult to maintain it during exploitation. Due to low hydraulic efficiency of the sediment tanks, unfavourable flocculation conditions in the combined flocculation tank and low water temperature,

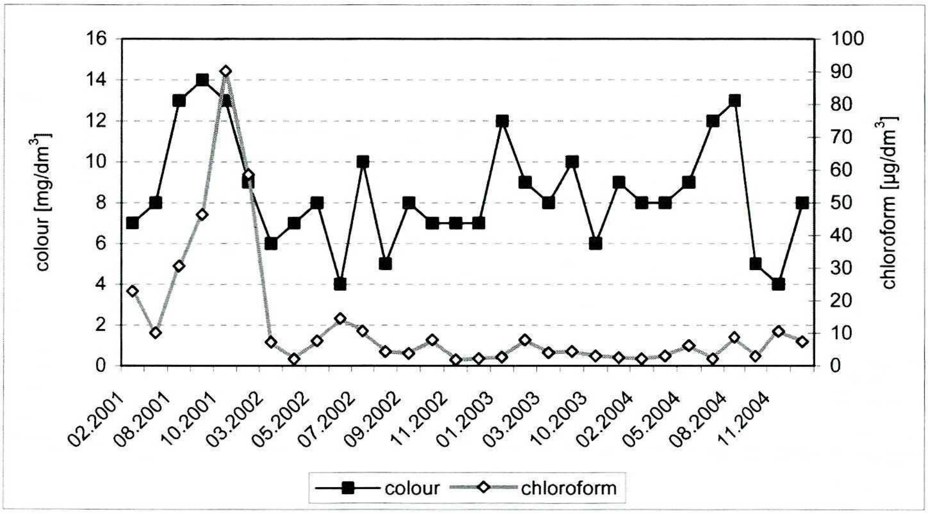

Fig. 7. Colour and chloroform in water after treatment

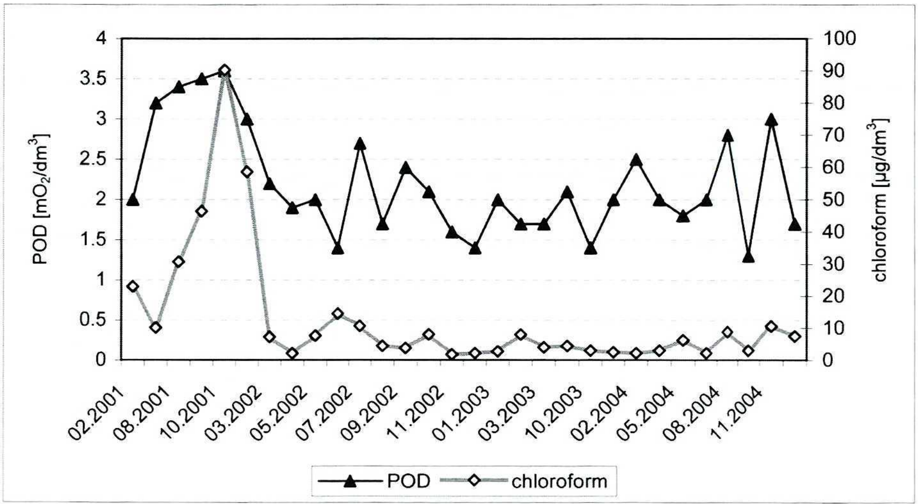

Fig. 8. POD and chloroform in water after treatment

improper coagulant dose (in aspect of water quality changes) caused delay in its hydrolysis. As a consequence, a decision was made to apply a new coagulant type. In laboratory research many hydrolyzing and pre-hydrolyzed coagulants were tested, in aspect of their effectiveness and the possibility of treatment without necessity of the pH adjustment. The crucial criterion of a reagent choice was its effectiveness in treatment of water of variable quality. Initially, a coagulant type and its dose was stated in jar testing. Then, a new coagulant was tested in the technical system. On the basis of those results Flokor 1,2 was chosen as the most effective coagulant and it was introduced in the plant without further pilot tests.

In the technical system dosing of Flokor 1,2A started in February 2007. In comparison to ALS treatment, "in-bed" coagulation with Flokor 1,2A was possible without the pH adjustment when water supplied to the WTP was of low turbidity. The average doses of pre-hydrolyzed reagent were ca. 2.0 mg Al/dm^3 in conventional coagulation and 0.7–1.3 mg Al/dm^3 in "in-bed" coagulation. The doses of pre-hydrolyzed coagulant were twice lower than ALS.

So that to assure stable technological system operation in aspect of raw water quality variations, some activities were undertaken, i.e. modernization of the rapid filters which involved a drainage system and exchange of sand bed for anthracite-sand bed. Treatment based on direct filtration resulted in decrease of reagents use and, what is especially important, effective DBPs precursors removal.

CONCLUSIONS

The water of the Wisła-Czarne reservoir is of very low hardness and low-mineralized. In spite of high SUVA values it is not susceptible to coagulation. The reason is probably lack of buffer capacity what requires the pH adjustment during coagulation. Another factor is low temperature of the water for many months in the year which inhibits the processes of coagulant hydrolysis and its precipitation.

The water requires treatment in "enhanced" coagulation because of the presence of dissolved organic matter which in disinfection with chlorine becomes a source of carcinogenic THMs in the amount exceeding even many times the level acceptable for drinking water.

To achieve the assumed results, the coagulation should be run in two optional technological systems – in conventional treatment and in direct filtration dependently of water quality and its temperature.

Effective treatment with ALS was possible, even at low temperature of the water, but at strict technological parameters (very low range of effective ALS dose and pH ~ 6). However, because of significant variations of water quality, it was very difficult to meet such strict requirements.

The application of pre-hydrolyzed Flokor 1,2A instead of ALS enabled to eliminate reagents to the pH adjustment and to apply "in-bed" coagulation when water supplied to the WTP was of low turbidity.

Treatment based on direct filtration with Flokor 1,2A and exchange of sand bed for anthracite-sand one ensured effective DBPs precursors removal.

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USUWANIE PREKURSORÓW UBOCZNYCH PRODUKTÓW DEZYNFEKCJI Z WÓD ZBIORNIKA ZAPOROWEGO

Wody ujmowane ze zbiornika zaporowego Wisła-Czarne należą do wód miękkich o niskiej zasadowości. Pomimo wysokich wartości SUVA należą do wód trudnopodatnych na oczyszczanie metodą koagulacji. W celu uzyskania założonych efektów uzdatniania proces koagulacji musi być prowadzony w różnych układach technologicznych (koagulacji objętościowej lub powierzchniowej), których wybór zależy od jakości oraz temperatury wody surowej. Skuteczne uzdatnianie badanych wód siarczanem glinu, nawet w okresach bardzo niskich temperatur wody było możliwe przy zachowaniu ściśle ustalonych parametrów technologicznych. Jednak z uwagi na dużą zmienność jakości ujmowanej wody, zwłaszcza w okresie intensywnych opadów atmosferycznych były one trudne do utrzymania. Zmiana rodzaju koagulantu na wstępnie zhydrolizowany koagulant glinowy Flokor 1.2A pozwoliła nie tylko na eliminację środków do korekty pH, ale także umożliwiła zastosowanie koagulacji powierzchniowej w okresach niskiej mętności wody surowej. Dzięki wprowadzeniu koagulacji powierzchniowej w połączeniu z modernizacją filtrów pospiesznych w zakresie konstrukcji drenażu oraz wymiany złoża z piaskowego na antracytowo – piaskowe uzyskano stabilniejszą pracę układu uzdatniania w aspekcie zmian jakości wody surowej, mniejsze zużycie reagentów oraz zapewniono pełne usuwanie prekursorów ubocznych produktów dezynfekcji.