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INFLUENCE OF SELECTED COAGULANTS OF INDICATOR AND DIOXIN-LIKE PCB REMOVAL FROM DRINKING WATER

WPLYW WYBRANYCH KOAGULANTÓW NA USUWANIE WSKAŹNIKOWYCH I DIOKSYNOPODOBNYCH PCB Z WODY PRZEZNACZONEJ DO SPOŻYCIA

Abstract: The aim of the research was to compare selected coagulants efficiency in indicator and chosen dioxin-like PCB removal from surface water. As coagulants, there were used aluminium sulfate and 5 hydrolyzed polyaluminium chlorides, with trade names: PAX-XL1, PAX-XL10, PAX-XL19, PAX-XL60, PAX-XL69. For the research, surface water was used, collected from dam reservoir. The water composition was modified with standard mixtures PCB MIX24 and MIX13, in order to obtain concentration of each congener equal to 300 ng/dm³. The PCB MIX24 mixture was composed of indicator congeners solution: 28, 52, 101, 118, 138, 153, and 180, whereas the MIX13 mixture - solution of three dioxin-like PCB 77, PCB 126, and PCB 169. It was demonstrated that the application of aluminium sulfate allowed for reaching better effects for purifying water of PCB, than with the usage of pre-hydrolyzed salts, polyaluminium chlorides. Out of the studied coagulants, the best effects for indicator PCB removal were obtained with the application of aluminium sulfate, total PCB concentration was decreased by 65%. The highest efficiency for indicator congeners removal (90%) was obtained for PCB 138 and 153. After the application of hydrolyzed polyaluminium chlorides PAX-XL1, PAX-XL10 decrease in higher chlorinated PCB concentration was obtained, in the range of 23 to 74%. Selectivity of chosen PCB congener removal, depending on applied coagulant, was demonstrated; with the usage of aluminium sulfate, removal of heptachlorobiphenyl PCB 180 at the level of 34% was obtained, whereas with the application of PAX-XL1 and PAX-XL10 higher reduction efficiency for this congener was obtained, i.e. 83 and 74% respectively. For dioxin-like PCB, after application of aluminium sulfate, total concentration reduction by 74% was obtained, efficiency of this congeners removal amounted to from 54 (PCB 77) up to 72% (PCB 126), similar results were obtained after the usage of PAX-XL1. The lowest PCB removal from water rate was stated for coagulants PAX-XL60 and PAX-XL69.

Keywords: polychlorinated biphenyls, coagulation, aluminium sulphate, hydrolyzed polyaluminium chlorides, removal efficiency, drinking water

Introduction

Contaminants present in water cause negative changes in physical, chemical, and bacteriological properties, which prevent the use of water for drinking and domestic purposes. These substances can be divided into several categories, one of which is

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classifications into organic micropollutants (e.g. polycyclic aromatic hydrocarbons, polychlorinated biphenyls, surfactants, pesticides), and inorganic micropollutants, primarily heavy metals [1, 2].

In recent years, there can be observed a significant increase in interest in organic micropollutants occurring in water at trace levels. These pollutants include e.g. endocrine disrupting compounds (EDCs), pharmaceutical and personal care products (PPCPs), disinfection by-products (DBPs), polychlorinated biphenyls (PCB), dioxins and furans (PCDF). Trace organic contaminants have become an increasing cause of concern for governments and water authorities around the world, as they attempt to implement sustainable water reuse practices [1].

PCB are harmful chemical compounds, however the degree of their toxicity depends on the amount and substitution position of chlorine atoms in a biphenyl molecule. 209 polychlorinated biphenyl congeners are known, a number of which are characterized by high bioaccumulation, toxicity or potential carcinogenicity, and simultaneously high persistence in the environment. So-called dioxin-like PCB, which include coplanar congeners with codes: 77, 81, 126, 169, are considered to be the most toxic. Three of them (PCB 77, PCB 126 and PCB 169) are spatial analogues of the most toxic dioxin, i.e. 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) [3]. According to the U.S. Environmental Protection Agency, 7 indicator congeners should be determined in the environment, with codes: 28, 52, 101, 118, 138, 153, and 180. Water ecosystems pollution occurs indirectly (from the atmosphere) or directly (discharge of waste water containing PCB, run-off from fields, leakages from transformers and condensers, landfills). Because of hydrophobic properties, PCB have a strong tendency to transition from liquid phase to phases with higher hydrophobicity, e.g. through bioaccumulation or sorption, by binding to sediment particles or to suspended solids present in water [4].

Negative influence of micropollutants on water consumers health causes the need to remove these substances from drinking water. The choice of micropollutant removal processes is determined by their type, properties, and form of occurrence. Therefore it is vital that effective water treatment processes are employed to remove trace organic contaminants, and thus ensure that any potential human health risks are mitigated prior to consumption [1].

Current literature reports emphasize the need for basic research regarding the removal mechanism of trace organic pollutants from water, using different processes. In the water treatment technology using chemical methods, the coagulation belongs to the most important unit processes. Its main purpose is to reduce turbidity and colour of water or wastewater, by degradation of colloid structures, intensification of flocculation of suspended solids, and intensification of sedimentation process of produced sediments [1, 5]. Moreover as a result of coagulation, decrease in concentration of organic substances, heavy metal ions, phosphates, silica, bacteria, and viruses occurs.

If organic compounds are present in water, usually precisely they will determine the conditions of the coagulation. This results among others from the fact that the surface charge of mineral particles is many times smaller than the charge of particles of organic substances, which are naturally present in water (NOM). In most natural waters with a typical range of pH, negative electric charge of mineral particles equals from 0.1 to 1 $\mu\text{eq}/\text{mg}$, while the charge of organic substances is usually 10 to 100 times higher. The presence of natural organic matter in treated water has an important impact on the coagulation process and on physical properties of the resulting agglomerates [1, 6].

Analyzing the mechanisms of removal of contaminants from water during the coagulation, it was found that for the removal of organic compounds mechanisms of complexation, neutralization and adsorption are responsible. However, which coagulation mechanism is dominant depends on many factors. According to the literature data the efficiency of the coagulation process is influenced by: the type of coagulant and its dose, the composition of purified water, including the content and properties of organic pollutants, pH, water alkalinity and physicochemical properties of removed organic micropollutants. One of these properties is the hydrophobicity expressed by *n*-octanol/water partition coefficient ($K_{o/w}$). The higher the $\log K_{o/w}$ value, the more insoluble in water the compound is, the easier and stronger it bounds to suspensions or sediment [7]. A relation between hydrophobicity of the micropollutants and the efficiency of the coagulation at a constant pH was demonstrated. For compounds with $\log K_{o/w} > 3.2$, high efficiency of their removal in the coagulation process was obtained. An example might be a research on the elimination of pesticides from surface water by means of coagulation and flocculation, which showed that the main mechanism regulating the removal of trace organic pollutants was sorption onto the natural material in the water [8]. Also Li et al. [9] studies confirm that during the coagulation of organic micropollutants, sorption of these compounds on the NOM particles plays an important role. The authors demonstrated that PCDD/F present in the water occur in a form bound to NOM, and during the coagulation they will be removed with the NOM particulates. The efficiency of PCDD/F removal in the coagulation process depends primarily on the volume and size of NOM particles, the type and dose of the coagulant and the pH. The dynamics of sorption of lipophilic organic micropollutants in water significantly affects their distribution in the two phases: suspended particulate matter (SPM) and water. The sorption and the desorption processes are regulated by the type of coagulant, and the mechanism of the coagulation process. The mechanism of the coagulation removal of NOM and hydrophilic micropollutants is mainly linked to the neutralization of colloidal NOM charge and precipitation as humates or fulvates, and coprecipitation by adsorption on humates or fulvates or on the metal hydroxide.

In order to remove the trace organic pollutants from water and wastewater, coagulants are usually selected based on the turbidity of the solution and physicochemical properties of the removed organic micropollutants. As coagulants, aluminium and iron salts are most often used. In recent years the use of new generation coagulants, with high efficiency, can be observed [1]. These coagulants reflect the properties of trace organic pollutants which are being removed (e.g. hydrophobicity, charge, polarizability, the presence of particular functional groups). It was also demonstrated that combined processes have a high efficiency in removal of organic micropollutants from water [9-11]. In the literature there are few examples of research regarding PCB removal from contaminated surface water. Due to the similarities in chemical properties of PCB and PCDD, for removing polychlorinated biphenyls from water, research developments in PCDD/Fs removal [9] can be used.

PCB pollution monitoring is particularly important for reservoirs, which are sources of drinking water for supplying the population. Research conducted in 2009-2011 for chosen water reservoir in Poland, which is the source of water for water treatment plant, showed that the water and the bottom sediments are contaminated with PCB [12]. The scope of PCB concentration in the reservoir water ranged from 1.0 to 8.1 ng/dm³, whereas the concentration of these compounds in the bottom sediments ranged from 0.12 to 2.78 µg/kg. The most commonly used coagulant in water treatment plants in Poland is aluminium

sulphate $\text{Al}_2(\text{SO}_4)_3$. Also, there are studies conducted regarding the application of pre-hydrolyzed coagulants, i.a. polyaluminium chlorides with general formula $\text{Al}_n(\text{OH})_m\text{Cl}_{3n-m}$. The mechanism of coagulation with aluminium salts, non-hydrolyzed and pre-hydrolyzed, is the same, however the presence of aluminium polymeric forms in polyaluminium chloride solutions cause them to be more stable in water, resulting in more efficient removal of pollutants [1].

The aim of this research was to compare the efficiency of chosen coagulants in removal of indicator and dioxin-like PCB from surface water.

Materials and methods

Research material

Kozłowa Góra water reservoir is created by damming the river Brynica. It is located on the south-eastern edge of Swierklaniec municipality. It occupies an area of approx. 5.5 km², its capacity equals to approx. 13 million m³, and its average depth is 4.5 m. Currently it constitutes water source for water treatment plant in Wymysłów, which belongs to Gornoslaskie Przedsiębiorstwo Wodociągów. The reservoir also fulfils flood prevention objectives, and it is used, to a limited extend, for touristic and recreational purposes.

Water from the reservoir, sampled in November 2011, was used for the research. Positions were situated in southern, outlet (by the dam) part of the reservoir. At each measuring point, 20 dm³ of water was collected. The water composition was modified in order to obtain concentration of each PCB congener equal to 400 and 300 ng/dm³ respectively, by introducing to water appropriate amounts of standard solutions - PCB MIX 13 and MIX 24.

As the coagulants there were used $\text{Al}_2(\text{SO}_4) \cdot 18\text{H}_2\text{O}$ produced by Przedsiębiorstwo Handlowe Polskie Odczynniki Chemiczne in Gliwice, and five hydrolyzed polyaluminium chlorides with trade names Kemira: PAX-XL1, PAX-XL10, PAX-XL19, PAX-XL60, PAX-XL69 produced by KEMIPOL in Police. Commercial solutions of polyaluminium chlorides were characterized by alkalinity equal to respectively 70±5%, 70±10%, 85±5%; 40±10%; 60±10%; 85±5%, and contained respectively 10.0±0.6%; 9.4±0.4%; 9.4±0.4%; 14.4±0.6%, and 11.3±0.9% of Al_2O_3 . For the research, 1% solution of aluminium sulphate, and solutions of polyaluminium chlorides were prepared, by diluting commercial products so that they contained 1.0 g Al/dm³.

Coagulation process

The coagulation process was conducted in glass vessels, to each 2 dm³ of studied water was measured. The coagulants were introduced in the amount of 4 mgAl/dm³, and fast stirring was performed for 2 minutes (applying 250 rpm) with the use of mechanical stirrer. Next slow stirring (20 rpm) was conducted for 15 minutes. After this time the samples were subjected to 1-hour sedimentation. Then 0.7 dm³ of water was decanted. Before and after the coagulation process, water analysis was performed, including determination of: pH, turbidity, colour, total organic carbon (TOC), and PCB analysis [12].

Analysis methodology

For the PCB analysis, hexane was added to 0.5 dm³ of sampled water, and it was stirred with magnetic stirrer. Next, after separation of hexane fraction in a separator, the solution was mixed with fresh batch of hexane. Hexane extracts were combined. The

extract was dried by seeping it through a layer of anhydrous Na_2SO_4 . Hexane was evaporated near to dryness from the dried extract under vacuum, and it was poured into a tube. The flask was further washed with hexane, and combined with the solution, and then transferred into the tube. For mineralization of organic compounds, obtained solution was shaken with concentrated H_2SO_4 . After separation of the mixture, 5% solution of KOH in ethanol was added to hexane layer. The tube was closed tightly and heated in a water bath. After cooling, ethanol-water solution was added (1:1, v/v). Next, the hexane layer was separated, which was concentrated in a vacuum evaporator to a volume of 1 cm^3 . All water samples collected in two measuring points underwent this procedure, in triplicate. The obtained extracts were analyzed qualitatively and quantitatively by means of capillary gas chromatography with mass spectrometry (CGC/MS). For the chromatographic analysis there were used standards by dr Ehrenstorfer company, i.e. PCB MIX 24 mixture, which was a solution of indicator congeners: 28, 52, 101, 118, 138, 153 and 180, and PCB MIX 13 solution, which contained three coplanar PCB congeners with codes: 77, 126 and 169 (Table 1). For their chromatographic separation, a DB-5 column was used. For detection, quadrupole mass spectrometer MS 800 by Fisons was used, which was operating in a selective ion monitoring mode. PCB quantification was achieved by single ion monitoring (SIM) [13].

Table 1

Composition of standard mixtures of indicator and dioxine-like PCBs

No IUPAC	Name of PCB	Concentration [mg/dm ³]	Pure [%]
Indicator PCB			
28	2,4,4-trichlorobiphenyl	10.0	97.0
52	2,2,5,5-tetrachlorobiphenyl	10.0	98.0
101	2,2,4,5,5-pentachlorobiphenyl	10.0	98.0
118	2,3,4,4,5-pentachlorobiphenyl	10.0	99.7
138	2,2,3,4,4,5-hexachlorobiphenyl	10.0	99.1
153	2,2,4,4,5,5-hexachlorobiphenyl	10.0	98.7
180	2,2,3,4,4,5,5-heptachlorobiphenyl	10.0	97.0
Dioxine-like PCB			
77	3,3',4,4'-tetrachlorobiphenyl	1.0	98.1
126	3,3',4,4',5-pentachlorobiphenyl	1.0	99.5
169	3,3',4,4',5,5'-hexachlorobiphenyl	1.0	99.7

In order to control and ensure the quality of the obtained results, prior to water samples analysis the performance and repeatability of the results were evaluated. As part of the performance evaluation, analysis of a blank sample, which did not contain PCB, was performed. In order to verify the repeatability, the water samples were subjected to triple, complete analytical procedure. The correctness of the approved methodology was verified by recovery determination. PCB extraction and qualitative-quantitative analysis were conducted parallelly according to the same rules in examined samples and in samples containing a known amount of standard mixture PCB MIX 24 or PCB MIX 13. PCB recovery ranged from 77 to 94%.

In order to evaluate the precision of the method, standard deviation for the obtained results was determined. The statistics of the significance of changes in PCB concentrations before and after the coagulation process was evaluated by *t*-Student t_d test too. This test was

used in order to verify whether there are statistically significant differences in average concentrations of PCB congeners before and after the coagulation process.

The degree of correspondence between the obtained results of PCB concentration in water samples after the coagulation process for individual coagulants ranged from ± 0.1 to ± 1.7 ng/dm³.

During the validation of the analytical procedure, based on determinations of the blank sample, basic level of noise was estimated, then the limit of detection (LOD) was determined and the limit of quantification (LOQ) was specified.

Results and discussion

Parameters of coagulation

The analysed surface water had slightly alkaline pH (7.9), colour equal to 30 mg Pt/dm³, and turbidity of 17 NTU [9]. Total concentration of indicator and dioxin-like PCB in sampled water was low and amounted to 32.3 and 4.7 ng/dm³ respectively.

Removal of PCB

Research results of PCB concentration in water after coagulation process for the coagulant dose of 4 mg Al/dm³ are presented in Figures 1 and 2.

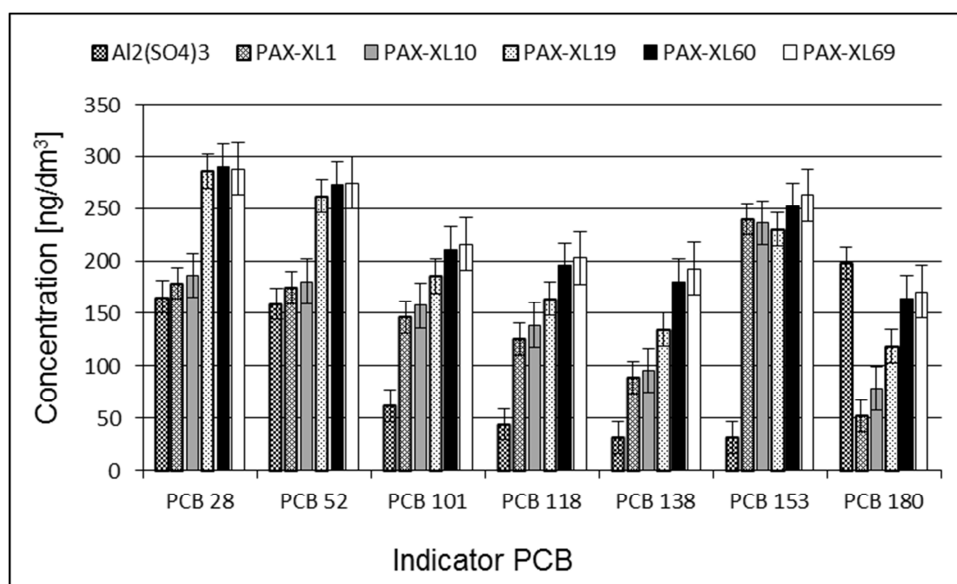


Fig. 1. Concentration of indicator PCB in water after coagulation process

Out of studied coagulants better effects of indicator PCB removal were obtained after the usage of aluminium sulphate, total PCB concentration decreased by 65%. The highest efficiency of indicator congener removal was obtained for PCB 138 and 153, which amounted to 90% (Table 2). The concentrations of other PCB were reduced in the range of 85 (PCB 118) to 34% (PCB 180). The LOD amounted to 0.1 ng/dm³.

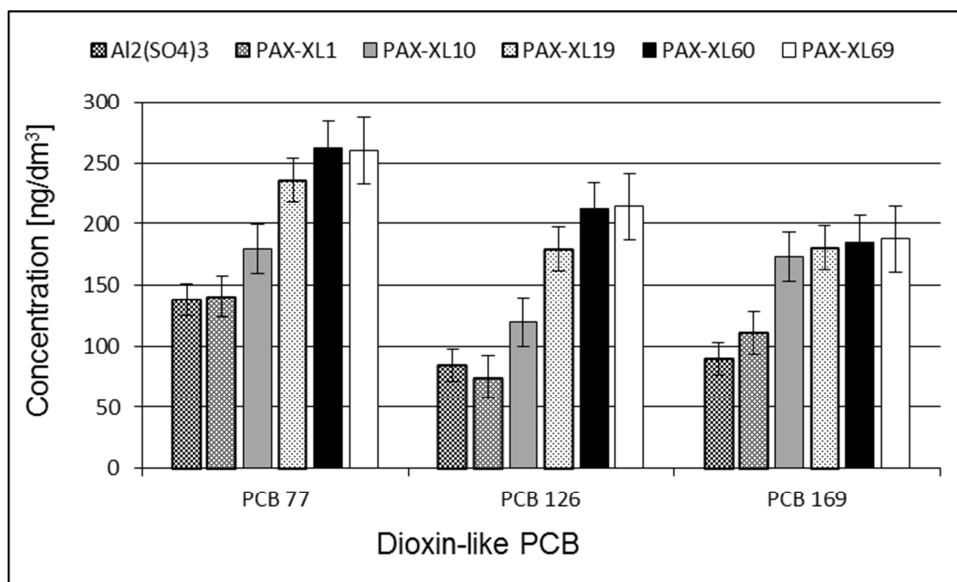


Fig. 2. Concentration of dioxin-like PCB in water after coagulation process

Table 2

Removal efficiency of indicator and dioxine-like PCBs from water after coagulation process

Congeners	Removal efficiency [%]					
	Al ₂ (SO ₄) ₃	PAX-XL1	PAX-XL10	PAX-XL19	PAX-XL60	PAX-XL69
Indicator						
PCB 28	45	40	38	5	3	4
PCB 52	47	42	40	13	9	8
PCB 101	79	51	48	38	30	28
PCB 118	85	58	54	45	35	32
PCB 138	90	70	68	55	40	36
PCB 153	90	21	21	23	16	12
PCB 180	34	83	74	60	45	43
Dioxin-like						
PCB 77	54	53	40	21	13	13
PCB 126	72	75	60	40	29	29
PCB 169	70	63	42	40	38	38

The effectiveness of aluminium sulphate in removal of hydrophobic organic pollutants (e.g. some of pharmaceuticals) was confirmed by research of Huerta-Fontela et al. [14]. Many authors emphasize the fact that in this process physico-chemical properties of removed contaminants are important [15], and the coagulant dosage, however that does not mean that the higher the coagulant dosage, the higher the pollutants removal efficiency, e.g. 78 mg/dm³ of aluminium sulphate dose was not effective for estrone and estradiol elimination (below 5%), whereas 50 mg/dm³ dose proved to be effective for phthalate plasticizers removal [16, 17].

Among hydrolyzed polyaluminium chlorides, the best results of indicator PCB removal were obtained after application of PAX-XL1 and PAX-XL10. For these

coagulants, decrease in total PCB concentration by 53 and 50% respectively was obtained. Greater reduction of higher chlorinated congener, i.e. PCB 180, 138 and 118, concentration was achieved, in the range of 54 to 83% (Table 2).

Selectivity in removal of chosen PCB congeners, depending on applied coagulant, was demonstrated; the usage of aluminium sulphate resulted in achieving heptachlorobiphenyl PCB 180 removal rate at the level of 34%, while using PAX-XL1 and PAX-XL10 resulted in obtaining higher reduction efficiency for this congener, i.e. 83 and 74% respectively.

For dioxin-like PCB after the application of aluminium sulphate total concentration reduction by 74% was obtained, congener removal efficiency amounted to from 54 (PCB 77) to 72% (PCB 126) (Table 2). Similar effects were achieved after the usage of PAX-XL1, total PCB concentration decreased by 73%, reduction of PCB 77, 126 and 169 concentration amounted to 53, 75 and 63% respectively.

The smallest degree of removal from water, both for indicator and dioxin-like PCB, was stated for coagulants PAX-XL60 and PAX-XL69. After the application of these coagulants, decrease in PCB concentration in the range of 3 to 45% (indicator PCB), and 13 to 38% (dioxin-like PCB), was obtained.

Obtained results confirm the relation between PCB hydrophobicity and the coagulation efficiency in removal of these contaminants from water. For all analysed coagulants, it was demonstrated that the higher $\log K_{ow}$, the higher the removal efficiency for indicator PCB in the range from PCB 28 to PCB 138. This correlation was not discovered for PCB 153 and 169, which suggests that the coagulation process is also affected by the structure of the compound [18], i.e. spatial position of chlorine atoms in biphenyl molecule (positions *ortho*-, *meta*-, *para*-).

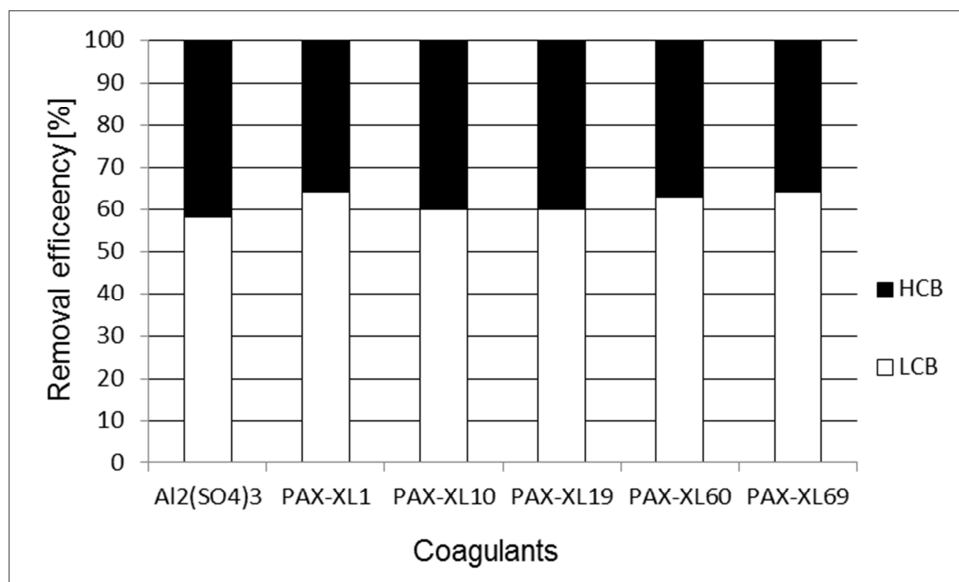


Fig. 3. Removal efficiency of LCB and HCB from water after coagulation process

The effectiveness of selected coagulants was also analyzed regarding the removal of lower and higher chlorinated PCB from surface water. According to literature data, the lower chlorinated PCB (LCB) include congeners containing up to four chlorine atoms in the biphenyl molecule, PCB containing more than four chlorine atoms in the molecule belong to higher chlorinated (HCB). For all tested coagulants, higher removal rate for LCB compared to HCB was obtained (Fig. 3). After applying hydrolyzed polyaluminium chlorides, LCB content in water decreased from 60 (PAX-XL10 and PAX-XL19) to 64% (PAX-XL1 and PAX-XL69). The coagulation with the usage of aluminium sulphate reduced LCB content by 58%, while HCB by 42%.

According to the literature data, the coagulation process with aluminium sulphate, apart from desirable effects, may cause negative changes in physico-chemical composition of purified water, e.g. an increase in aluminium ion concentration or intensification of water corrosivity. Increased aluminium ion concentration in drinking water may also pose a potential threat to human health [1]. Therefore, despite demonstrated in the research, higher efficiency of aluminium sulphate in PCB removal from water, it is suggested to use polyaluminium chlorides PAX-XL1 and PAX-XL10, which have alkalinity equal to $70 \pm 5\%$, $85 \pm 5\%$ respectively, and which contain approx. 10 and 3% of Al_2O_3 . The application of polyaluminium chlorides as coagulants for organic pollutant removal, is recommended by many authors [1, 2]. Higher efficiency is explained by the fact that polyaluminium chloride solutions contain more polycationic and polymerized products of controlled prehydrolysis than aluminium sulphate. In the pH range of natural waters and with their alkalinity, hydrolysis of aluminium cations present in aluminium sulphate occurs almost immediately after their contact with treated water, therefore the precipitation of aluminium hydroxide occurs faster than the desired reaction of colloids and organic pollutant anions neutralization [2]. The obtained results suggest that in case of removal from water indicator and dioxin-like PCB using polyaluminium chlorides, their alkalinity is important, which should amount to approx. 70%. Alkalinity of PAX-XL60 and PAX-XL69 was lower and amounted to approx. 40 and 60%, which can explain lower efficiency of these coagulants in PCB removal. PAX-XL1 coagulant efficiency in congener removal is confirmed by results obtain for dioxin-like PCB. Also the literature data confirm the effectiveness of polyaluminium chloride in dioxin and dioxin-like compound removal. Li et al. [9] demonstrated 99% efficiency in PCDD/Fs removal after application of ferric chloride and polyaluminium chloride, slightly lower (97-98%) was obtained for aluminium sulphate. Applied coagulants were selective in removal of chosen PCDD/Fs. The literature data and own research indicate that significant improvement of polyaluminium chloride efficiency in PCB removal from water is achieved by application of coagulation process enhancement with powder-activated carbon. Liyan et al. [19] demonstrated the effectiveness of powder-activated carbon (PAC), granular-activated carbon (GAC) in hydrophobic organic chemicals (HOCs) removal, in the range of 73.4 to 89.2%, which is the next stage of own research.

Conclusions

Conducted research allowed for formulating the following conclusions:

- good effects of indicator and dioxin-like PCB removal from surface water were obtained after the application of aluminium sulphate, and hydrolyzed polyaluminium chlorides PAX-XL1 and PAX-XL10,

- after the coagulation process using aluminium sulphate and coagulants PAX-XL1 and PAX-XL10, total concentration of indicator PCB decreased in water by 65, 53, and 50% respectively,
- for dioxin-like PCB, after using aluminium sulphate and hydrolyzed polyaluminium chloride PAX-XL1, total concentration reduction by 74 and 73% was achieved,
- for the analyzed coagulants, it was demonstrated that with the increase in PCB hydrophobicity, the efficiency of removal of these compounds increase in the range from PCB 28 to PCB 138,
- after the coagulation process, for all of the analyzed coagulants, higher elimination rate for lower chlorinated congeners was obtained, compared to higher chlorinated PCB,
- selectivity in chosen PCB congener removal was demonstrated, depending on applied coagulant; with the usage of aluminium sulphate removal of heptachlorobiphenyl PCB 180 at the level of 34% was obtained, whereas with the application of PAX-XL1 and PAX-XL10 higher efficiency of this congener reduction was obtained, i.e. 83 and 74% respectively.

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