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Comparative studies of coagulation in a batch and continuous mode systems

Marek Mołczan[1,2]*, Małgorzata Wolska[1,2], Sławomir Szerzyna[1], Małgorzata Żółtowska[2], Jacek Wiśniewski[1], Wojciech Adamski[1]

[1]Wrocław University of Science and Technology, Poland
Faculty of Environmental Engineering
[2]Municipal Water and Sewage Company in Wrocław, Poland
Center of New Technologies

*Corresponding author's e-mail: marek.molczan@pwr.edu.pl

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Abstract: Coagulation studies were carried out simultaneously in a batch and continuous mode systems. Four coagulants of varied properties were used. Both test procedures contained the same process phases (rapid mixing, flocculation, sedimentation) and a corresponding coagulant dosage range (1.7–3.3 gAl/m^3). In the batch mode test, the raw water composition was stable, while in the continuous mode test it was subject to changes resulting from natural variation in source water properties. The main process effectiveness measures were: color, absorbance at 254 nm, and dissolved organic carbon content. Studies in the batch mode system showed clear linear relationships between coagulant dosage and effectiveness of analyzed water contamination indicators values reduction. Tests in flow conditions confirmed the relationships found in the batch test, albeit with lesser agreement with a linear effectiveness – coagulant dosage model. Due to a comparison of coagulation results from both test systems, it was possible to determine the relationships between coagulation effectiveness obtained in continuous and batch mode systems. The linear relationships obtained show a dependence on current raw water properties and the type of coagulant used. However, in many cases, these relationships do not differ significantly as with respect to analyzed water quality indicators. The general rule is that the effectiveness observed in the continuous mode system is larger than that for the batch mode process, which is justified due to different process conditions. Determining the relationship between coagulation results obtained for different experimental procedures allows for predicting effects of changes in technical systems based on laboratory results.

Introduction

Coagulation is doubtlessly a key surface water treatment process. The coagulant type and dosage are the main process tools (Sillanpää et al. 2018). The coagulant dosage value may be connected with properties of raw water and the treated water quality or the degree of colloidal system destabilization (Ratnaweera and Fettig 2015). In the first case, in order to evaluate water contamination, turbidity (an indicator of undissolved mineral content) and indicators of organic substance content (color, total or dissolved organic carbon, UV absorbance) are taken into account. In the second case, the residual coagulant content and factors connected with water disinfection are additionally taken into account (Dąbrowska et al. 2016). In the last case, the zeta potential or its surrogate in the form of stream current are evaluated (Kłos et al. 2011, Kłos 2012).

The commonplace use of coagulation, significant variation in content and properties of raw water and large choice of coagulants lead to the fact that coagulation conditions are subject to frequent revision. A commonly applied procedure used to this end is a jar test (Manual M37 2011). This is conducted at well-defined and, above all, repeatable conditions. Sedimentation is carried out in the same reactor as previous phases of colloid destabilization and flocculation. The water content and temperature are stabilized. Coagulant dosage is precise and is not subject to disturbances caused by variations in water flow rates. Almost all of the process parameters can be freely changed and rigorously maintained.

The conclusions derived from experiments in a batch mode system are then used for operating the process at flow conditions, which are significantly different. This applies not only to qualitative and quantitative variation in raw water. A significant factor is also the characteristics of the hydraulic system. The individual reactors are usually separated. Their dimensions determine the relationships between values of retention times for individual process phases, which therefore cannot be arbitrarily set. The mixing conditions are determined not only by stirrer motion but also by hydrodynamic effects connected with water flow through the reactor. The system of pipes connecting the tanks containing individual process phases is also of importance. Even if criteria concerning the flow speed regime are fulfilled, then the piping geometry, mounted fittings and similar factors, to a greater or lesser degree, favor hydraulic impulses, gas accumulation, precipitate accumulation and coagulation floc break up.

Finally, the effects of coagulation in a continuous mode system are the result of chemical and physical transformations of coagulation substrates caused by interaction with the coagulant and complex process operating conditions. The complexity of these conditions is significantly greater in this case than in a batch mode system. Despite these differences, many years of operating experience indicate that finding a relationship between the process effects in batch mode and flow type systems is possible. Seeking the nature of this relationship, on the basis of small-scale batch and continuous flow laboratory systems, was the main purpose of the study.

Experimental section

Test procedures

In the initial stage of the study, four coagulants out of ten tested were chosen. The choice was not only based on contamination removal effectiveness, but above all on variation in properties and purchase costs (C1<C3<C2<C4). At this stage the dosage ranges were determined, establishing limits where a change in dosage yields clear changes in contaminant removal effectiveness, and with the ranges being within the limits of economic viability.

Studies in the flow type system (100 dm^3/h) were conducted in an experimental system equipped with rapid and slow mixing chambers and a lamella plate clarifier with concurrent flow pattern (Fig. 1). The studies were carried out in four cycles (4 x 5 days, four weeks in late July and early August). In each cycle, a different coagulant was tested, and during each day a different coagulant dosage was used. For each dosage, three series of measurements were performed for both raw and treated water. The first series of measurements took place after about 16 hours of continuous system operation to allow for operating conditions to stabilize. Subsequent measurement series were performed at two hour intervals, after which the change in dosage/coagulant type was made to perform another 24 hour measurement cycle. Treated water samples were taken with a delay relative to raw water samples resulting from the water retention time in the treatment system (40 minutes). The base for agreement in results between batch and continuous mode systems was taken to be the coagulant type and dosage, as well as flocculation conditions. The flocculation time in a continuous mode system was 19 minutes, and the sedimentation time about 20 minutes. The relatively short sedimentation time was of secondary importance, since only indicators of dissolved substance content were used for evaluating process effectiveness. The mixing intensity was selected experimentally based on observation of floc formation and were 200 rpm and 40 rpm for rapid and slow mixing respectively. Minimal mixing intensity which caused whole reactor volume opacification with coagulant dosing, within no more than 60 seconds, were chosen for fast mixing. As well minimal mixing intensity which caused flocs formation without sedimentation was chosen for flocculation step.

For each coagulant, a jar test was performed in the full range of dosages used in the continuous mode system (1.7–3.3 gAl/m^3). The process was performed with the use of a laboratory flocculator and glass beakers filled with 1 dm^3 water samples. Coagulant dissolution was performed for 2 minutes at a stirrer speed of 150 rpm. During flocculation (20 min), the stirrer speed was reduced to 20 rpm. Mixing intensity parameters were established with the same criteria as for continuous mode system. The samples then underwent sedimentation for one hour. Analysis was performed on raw water and water decanted after sedimentation.

Materials

Coagulation was performed on water from the Oława and Nysa Kłodzka rivers, the sourced for water supply purposes of the city of Wrocław. Its characteristics, with respect to all measurement cycles, are shown in Table 1.

In this study four coagulants were used whose characteristics are summarized in Table 2. All were aluminum coagulants. Aluminum sulphate (C1) was the simplest structurally.

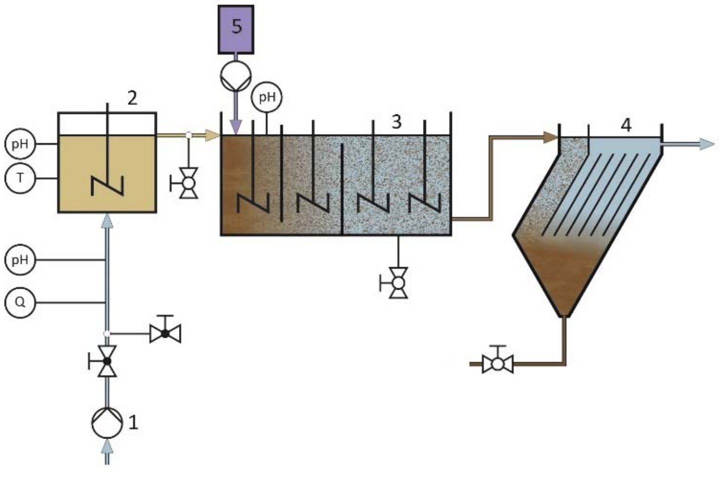

Fig. 1. Schematic of the test flow system with supply pump (1), raw water tank (2), flocculation chamber (3), settling tank (4), system for dosing coagulant into the rapid mixing chamber (5) and flow (Q), pH, and temperature (T) measurement points

Subsequent coagulants (C2-C4) were polyaluminum chlorides, differing mainly due to their metal content (5.3–10.5%) and degree of hydrolization (basicity) – ranging from 37% (C3) through 70% (C2) to 85% (C4). The applied doses of coagulant were homogenous in terms of metal (aluminum) content. The aim of this study was not comparing the effectiveness of coagulants but to verify if and to what degree the type of coagulant used influences the relationships between results obtained in batch and continuous mode systems.

Apparatus and analysis

This study made used of the following analytical apparatus: a Hach HQ40d multiparameter meter (temperature, pH, conductivity), a Shimadzu UV-1800 spectrophotometer (color at 340 nm and 410 nm, UV absorbance at 254 nm), a Hach 2100AN turbidimeter (turbidity) and a HiperTOC Thermo Scientific total organic carbon analyzer, working in an oxidation with ammonium persulfate mode (DOC). Alkalinity was determined by titration according to PN-EN ISO 7887:2012. Samples for spectrophotometric and dissolved organic carbon (DOC) analysis were filtered through a 0.45 µm membrane filter. All analyses were performed according to Polish standards.

The performance of color analysis at wavelength of 340 nm and 410 nm results from the provisions of Polish Standard PN-EN ISO 7887:2012, which allows for the determination of

Table 1. Variation in raw water parameters used in tests with coagulants C1-C4 in the batch mode system (BM) and flow type system (FS)

Test / Parameter	C1 (BM) C1 (FS)	C2 (BM) C2 (FS)	C3 (BM) C3 (FS)	C4 (BM) C4 (FS)
Temperature,°C	24.1	21.0	22.6	25.6
	22.6–25.3	20.8–22.3	21.0–24.2	23.3–25.4
pH, -	7.55	7.86	7.74	7.85
	7.34–7.53	7.38–7.63	7.56–7.81	7.62–7.97
Conductivity, µS/cm	372	370	369	354
	368–375	364–375	360–372	346–361
Alkalinity, $gCaCO_3/m^3$	122	117	116	114
	115–122	116–121	112–120	108–114
Turbidity, NTU	16.5	15.6	22.1	8.7
	1.3–28.7	1.4–30.7	9.0–48.4	5.3–27.6
Color 410 nm, gPt/m^3	15.5	17.6	20.5	18.4
	10.2–17.5	16.7–19.0	18.2–24.2	17.4–19.3
Color 340 nm, gPt/m^3	9.0	10.0	10.6	9.8
	8.4–10.1	9.7–10.3	9.8–11.4	10.0–10.5
Absorbance 254 nm, m^{-1}	9.8	10.2	10.7	9.2
	9.3–10.5	9.6–10.5	10.2–11.0	10.2–11.6
DOC, gC/m^3	4.6	4.6	4.6	4.4
	3.7–4.8	3.7–4.4	4.1–5.0	4.1–4.8
SUVA, $m^3/gC \cdot m$	2.1	2.2	2.3	2.1
	2.1–2.6	2.3–2.7	2.1–2.6	2.3–2.6

Table 2. Characteristics of coagulants used in the study

Coagulant code	C1	C2	C3	C4
Commercial name	ALS	PAX XL3	PAX 16	PAX XL1910
Al^{3+} content,%	4.2 ±0.2	5.3 ±0.3	8.2 ±0.2	10.5 ±0.5
Cl^- content,%	–	13.0 ±2	19 ±2.0	6.0 ±0.5
Basicity[1),]%	–	70 ±5	37 ±5	85 ±5
pH, –	2.4 ±0.5	2.5 ±0.5	1.0 ±0.2	4.0 ±0.5
Density, g/dm^3	1310 ±10	1210 ±40	1330 ±20	1280 ±30
Modifiers	–	Na^+	–	–

[1)] The percent of coagulant basicity represents the portion of acidity of alum that has been preneutralized.

color at 410 nm (recommended by sanitary services) as well as at conditions of maximum absorbance observed for specific natural water source (in this case 340 nm).

Due to non-standard sedimentation conditions in the continuous flow system (only 20 minutes settling time in order to retain a flocculation time similar to that in the batch mode system), data concerning turbidity are not presented, despite being collected during the study.

Results and discussion

The studied water was characterized by a relatively low susceptibility to treatment with the coagulation process, which is shown by an average organic substance content (Table 1), the water alkalinity value (about. 110–120 $gCaCO_3/m^3$) and a small value of specific UV absorbance (2–3 $m^3/gC\cdot m$) (US EPA 1998, Edzwald and Tobiason 1999, Volk et al. 2000, Machi and Mołczan 2016).

The variability in the characteristics of the source water (Table 1) well illustrates one of the main differences between coagulation conditions in batch and continuous mode systems. For example, raw water turbidity in the batch mode system with coagulant C3 was 22 NTU, while during a continuous flow test conducted for five days it changed in the range of 9–48 NTU (Table 1). However, in majority of cases, the values of raw water quality indicators in the batch mode system were contained in the range observed for the continuous mode system, which testifies that the quality of water used for the batch mode test is representative of the flow system.

In the range of coagulants and dosages used, linear relationships were found between the coagulant dosage and the effectiveness of decreasing the analyzed water contamination indicators in the batch mode system (Fig. 2). The values that were found for the linear determination coefficients show that the linear nature of these relationship is not accidental (in many cases $R^2>0.99$). The nature of this relationship, resulting from the mechanisms of the coagulation process, should be maintained when conducting the process at flow conditions.

Regardless of the coagulant used, its dosage and the variability in source water composition, the effectiveness in reducing the values of individual parameters is characterized by the series: color at 410 nm > color at 340 nm > UV_{254} > DOC. Only in the case of coagulant C4 similar effectiveness values were found for DOC and absorbance at 254 nm.

Studies on flow conditions confirmed the relationships found in the batch mode system, but with a smaller correspondence of the results with the linear relationship between process effectiveness and coagulant dosage (Fig. 3). Out of 16 series of data, only for four the determination coefficient was $R^2>0.9$, in eight cases $R^2>0.8$, and in two $R^2<0.5$. The rule, however, was a greater removal effectiveness in the continuous mode system. There is no clear evidence, but it is possible that the cause lies in the scale of the installation and in the movement of the liquid itself, which in the continuous mode system occurs not only during flocculation but also during sedimentation step. A larger reactor potentially allows for greater potential of interaction between agglomerate particles and dissolved pollutants. Furthermore, reactions connected with this do not stop at the stage of suspension separation in the settling tank, where the floccules still have the possibility to grow and contact the counterflowing water. It must be noted that the difference in scale causes more intense gas exchange and a faster transfer of carbon dioxide into the atmosphere in the laboratory scale batch mode reactor (Letterman and Yiacoumi 2011). Therefore, larger reactors are characterized by a lower post-coagulation pH value, which generally increases the process effectiveness (Świderska et al. 2008).

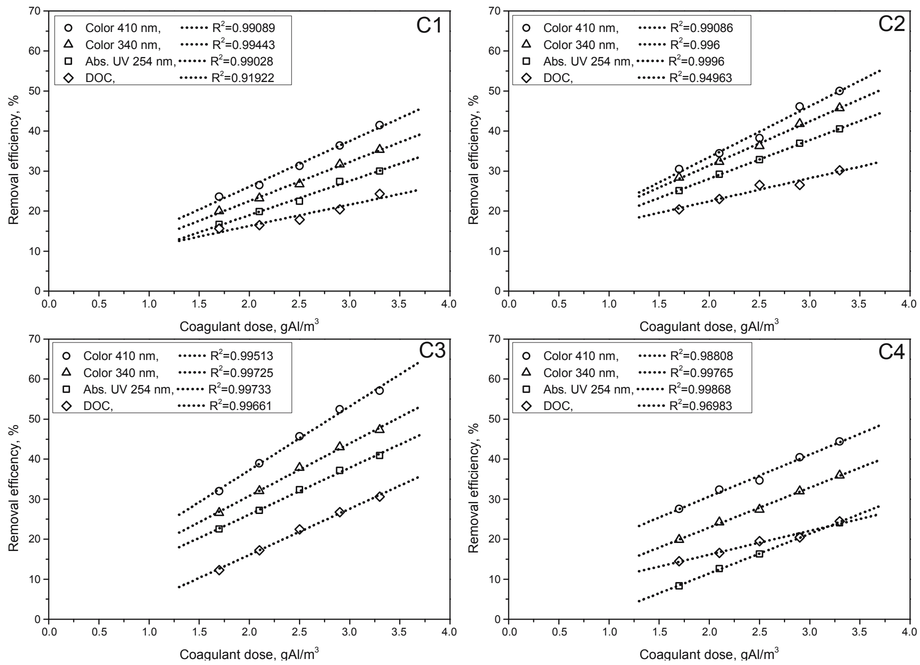

Fig. 2. Effect of coagulant dosage on coagulation effectiveness with C1-C4 coagulants for the batch mode system

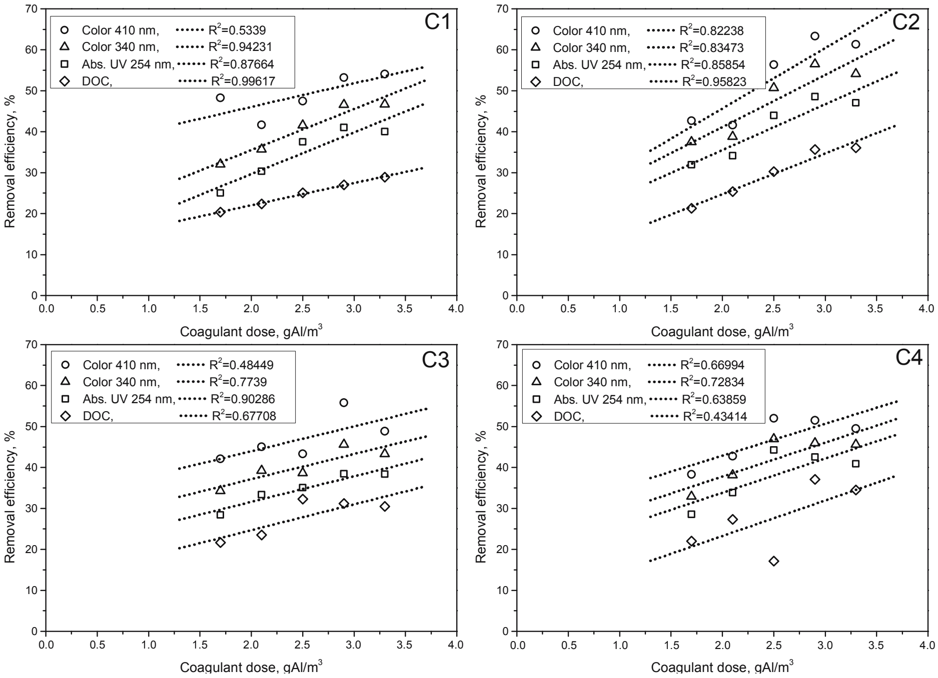

Fig. 3. Effect of coagulant dosage on coagulation efficiency with coagulants C1-C4 in the continuous mode system

A linear equation in slope–intercept form has two factors, which may be subject to interpretation. The intercept shows the reaction of the treated medium on introduction of coagulant (the greater value illustrates more intense reaction). On the other hand, the slope value indicates the sensitivity of the reaction to increases in coagulant dosages. In this aspect, the results from the batch and continuous mode were not completely coincidental. Whereas in the batch test the greatest sensitivity to coagulant dosage was found for coagulant C3, in the flow study this was true for coagulant C2. Such effects may also be connected with different process conditions characteristic to given reactor system.

The linear relationship between coagulation effectiveness and coagulant dosage, observed particularly clearly in the batch system, also allows for finding a linear relationship directly connecting the results of tests in batch and continuous mode systems (Fig. 4). Agreement with the linear relationship is in this case similar to that obtained for the continuous mode system. The character of this relationship indicates that, as expected, it will be dependent of the current characteristics of treated water and the type of coagulant used. On the other hand, it may be somewhat independent of quality indicators of water undergoing treatment. This may be clearly seen with respect to results from coagulants C2 and C3. In the case of remaining coagulants, the color indicator at 410 nm for coagulant C1, and DOC along with, to a limited degree, UV at 254 nm absorbance (slope value similar to that for other indicators) for coagulant C4 are the exceptions (Fig. 4).

Due to differences in composition of water undergoing treatment with individual coagulants, a direct comparison of their effectiveness is not possible. Based on the results obtained, it may be concluded that the relationship between coagulation effectiveness in the batch and continuous mode studies may be specific to a given coagulant. This results from, among other things, different effectiveness ranges found for individual coagulants, where the differences in raw water composition were not significant, e.g. with respect to coagulant C1 and C2 (Fig. 4). It is a well-known fact that besides NOM properties (Su et al. 2017) there are also several properties of coagulants (e.g. degree of polymerization, basicity, etc.) that may cause diverse impacts on organics removal efficiency (Liu et al. 2009, Yang et al. 2011, Lin et al. 2014, Dąbrowska 2016, Zhang et al. 2017). Therefore, coagulants of broadened spectrum of activity, like ion exchange coagulants (Zhao et al. 2016), might play the special role in the future.

Color is a unique water quality indicator, because it may be directly evaluated by the consumer, yet it does not have a clearly specified limit value (MH Regulation 2017). Various methods of color measurement are not however in agreement with the values obtained. Measurement of raw and coagulated water conducted for wavelengths of 410 nm and 340 nm have shown that color values at 410 nm were always greater. At the same time, a strict linear relationship was found between these two water color values. However, linear relationships between the two indicators were different for raw and coagulated water. This relationship may also change in subsequent water treatment stages. The choice of color measurement method therefore is of importance in evaluating the effectiveness of subsequent water treatment stages.

Several jar test experimental procedures could be used for various coagulation parameters optimizing (Ramphal and Sibiya 2014) but batch method results will never be in full conformity with continuous flow system effects. For technical unit operation (with fixed size of reactors and agitators) coagulant type and dosage are key operating parameters. For specified coagulant and determined raw water composition jar test treatment efficiency could be related to flow system efficiency. Performed research indicates that it could be

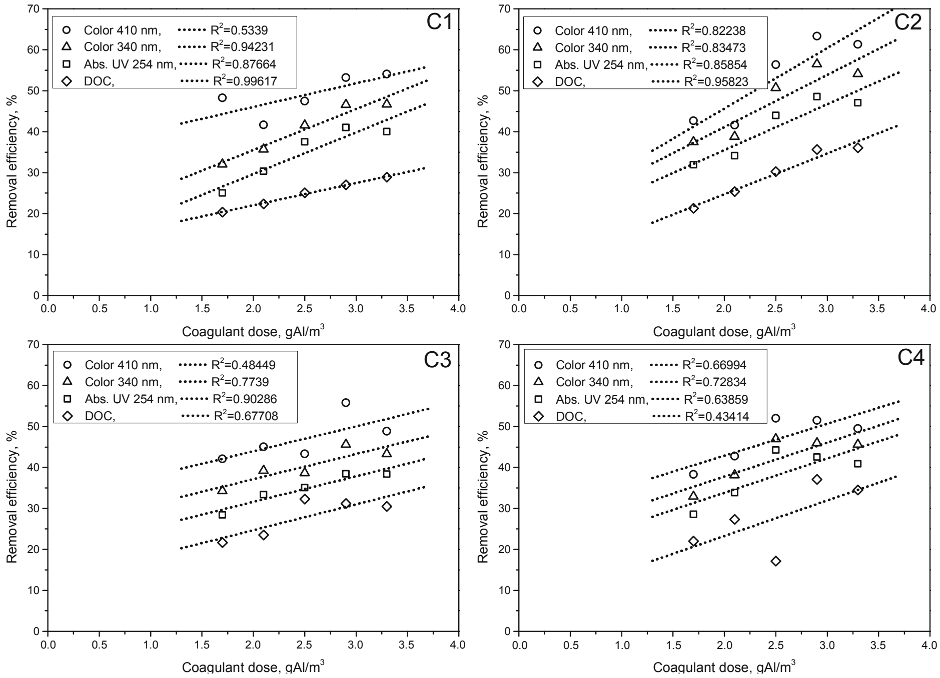

Fig. 4. Relationship between coagulation effectiveness in continuous mode and batch mode systems

a linear relation. Batch method is not a treatment option but it is often exploit solution helpful to setting technical scale flow mode system parameters. The research had a case study character, but compared with other literature reports could also be used for the future evaluation relationships of more general sense.

Conclusions

In the studied coagulant dosage range a clear linear relationship was found between the dosage and coagulation effectiveness. Due to the variation in process determinants, typical for continuous mode systems (especially with respect to raw water quality), the linear character of this relationship was more visible in results from the batch mode system. By comparing the coagulation results in both test systems it was possible to find the relationships between the effectiveness found in batch and continuous mode systems. The resulting relationships show a dependence on the characteristics of water being treated and the type of coagulant used. However, in many cases these relationships do not differ with respect to the analyzed water quality indicators. The general rule is that the effectiveness of coagulation observed in the continuous mode system is greater than that observed in the batch mode system.

It should be noted that the research flow reactor differs both structurally and hydraulically from devices on a technical scale. As a batch mode system is an imperfect process model, so is a laboratory flow system only similar to technical-scale systems. Nevertheless, due to the impossibility of conducting experiments directly in installations treating water for human consumption, every possibility of predicting the operating effects of such installations based on available research methods is valuable.

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Badania porównawcze koagulacji w układzie porcjowym oraz przepływowym

Streszczenie: Prowadzono jednoczesne badania koagulacji w układzie porcjowym oraz przepływowym. Korzystano z czterech koagulantów o zróżnicowanych właściwościach (polimeryzacja, zasadowość, zawartość glinu, koszt zakupu). Obie procedury badawcze zawierały te same fazy procesu (szybkie mieszanie, flokulację, sedymentację) oraz analogiczny zakres dawek koagulantów (1,7–3,3 gAl/m^3). W teście porcjowym skład wody surowej był ustalony natomiast w badaniu przepływowym podlegał zmianom wynikającym z naturalnej zmienności charakterystyki ujmowanej wody powierzchniowej. Głównymi wskaźnikami oceny skuteczności procesu były: barwa, absorbancja w 254 nm oraz zawartość rozpuszczonego węgla organicznego. Badania w układzie porcjowym pokazały wyraźne liniowe zależności wiążące dawkę koagulantu ze skutecznością zmniejszania wartości analizowanych wskaźników zanieczyszczenia wody. Badania w warunkach przepływowych potwierdziły prawidłowości odnotowane w układzie porcjowym, jednak przy mniejszej zgodności uzyskanych wyników z liniowym modelem zależności skuteczności procesu od dawki koagulantu. Dzięki zestawieniu wyników koagulacji w obu układach doświadczalnych możliwe było określenie związków skuteczności koagulacji osiąganych w układzie przepływowym i porcjowym. Uzyskane liniowe przebiegi wykazują zależność od aktualnej charakterystyki oczyszczanej wody oraz rodzaju stosowanego koagulantu. Natomiast w wielu wypadkach nie różnią się istotnie w odniesieniu do analizowanych wskaźników jakości wody. Za regułę uznać można większą skuteczność koagulacji obserwowaną w układzie przepływowym wobec układu porcjowego, co znajduje uzasadnienie w różnych warunkach prowadzenia procesu. Ustalanie związków pomiędzy wynikami koagulacji uzyskiwanymi w różnych procedurach badawczych, a w szczególności w układzie porcjowym oraz przepływowym, pozwala na przewidywanie skutków zmian wprowadzanych w układach technicznych w oparciu o efekty doświadczeń laboratoryjnych.