Relationships between floc features and coagulation-flocculation treatment efficiencies of various model wastewaters

Y. Kalashnykov[1], N. Sivchenko[2]*, H. Ratnaweera[2], K. Kvaal[2]

[1] Ukrainian State University of Chemical Technology, 49005, Gagarin av., 8, Dnipropetrovsk, Ukraine
[2] Norwegian University of Life Sciences, PO Box 5003-IMT, 1432 Aas, Norway
* Corresponding author. E-mail: nasi@nmbu.no; tel.: +47 64 96 64 23

Abstract

Coagulation is a widely used process for transformation of small particles into larger aggregates with further removal of these impurities by sedimentation, flotation, filtration, centrifugation or other separation methods. Study of the relationships between wastewater, coagulants, operational properties, features of aggregates and resulting efficiencies provides valuable input to the modelling of coagulation process and could be used for coagulant on-line dosage control. Experiments were performed to ensure that using images of flocs it is potentially possible to predict the optimal dosages of coagulants for each particular type of model wastewater and predict the treatment efficiencies. Preliminary tests showed that all coagulants have similar trends regarding treatment efficiencies for different model wastewater types during coagulation with a constant pH. Thus, three coagulant doses were chosen to analyse relationships between floc features, doses and treatment efficiencies for various model wastewaters and three coagulants. Floc features in this study are represented by image texture analysis – GLCM. Obtained results show the correlation between coagulant doses, treatment efficiencies and GLCM texture parameters. GLCM texture parameters of floc images tend to be unique for optimal dosages of coagulant.

Аннотация

Коагуляция – это часто используемый процесс укрупнения частиц с их последующим извлечением посредством осаждения, флотации, фильтрации, центрифугирования или другими методами раздела фаз. Изучение взаимосвязей между параметрами сточной воды, коагулянтами, рабочими параметрами процесса, характеристиками частиц и полученными степенями очистки от загрязнителей является важным этапом в моделировании процесса коагуляции, и в последующем может быть использовано для контроля дозы коагулянта в режиме онлайн. Проведена серия экспериментов с целью подтвердить, что анализ изображения хлопьев – это метод, который потенциально может быть использован для определения оптимальной дозы коагулянта и предсказания степени очистки для каждого конкретного вида модельной сточной воды. Предварительный анализ показал, что все коагулянты имеют схожие тенденции касательно степеней очистки от разных загрязнителей для разных типов модельной воды при постоянном pH во время коагуляции. Таким образом, было выбрано три дозы коагулянта для последующего изучения взаимосвязей между характеристиками хлопьев, дозами коагулянта и степенями очистки для различных типов модельной воды и трех коагулянтов. Характеристики частиц в этом анализе определяются с помощью текстурного анализа изображений – GLCM. Полученные результаты исследования подтверждают взаимосвязь между дозами коагулянта, степенями очистки от загрязнителей и текстурными параметрами GLCM изображений хлопьев. Для оптимальных доз коагулянта текстурные параметры GLCM изображений хлопьев являются уникальными.

Keywords: Optimization of coagulation; image analysis of flocs; GLCM of floc images; dosage prediction; efficiency prediction; model wastewater.

Introduction

Wastewaters contain a significant amount of suspended and dissolved substances, which create the need for its treatment before discharging into the receiving waterbodies. It is well known that insufficient wastewater treatment could dramatically influence on the environment (Smith et al, 2009; Anderson et al, 2002). It causes algal blooms that lead to death of fish and vegetation and enhances bacteria growth (Corell, 1998). These factors can become sources of serious health problems such as dysentery and cholera (Lipp et al, 2002). Thus, the quality control at water and wastewater treatment plants should be as high as possible.

At first treatment plants the quality control was performed by manual water sampling after certain stages of treatment (Edzwald, 1999). The main problem of this approach was a very long response time from sampling until results obtaining. This leads to usage of large amounts of chemicals in order to preempt the possible water quality problems. To eliminate mentioned disadvantages current W&WWTPs are using water quality control based on on-line water parameter analysis (Vesilind, 2003; Bourgeois et al, 2001). In this approach the sampling and analyzing of water parameters are typically automatized while process control is carried out by on-line monitors coupled with optimal coagulant dosing control systems (Ratnaweera, 2004). However, initial costs of such monitors are quite high.

The main objective of this research was to find a simple tool to predict the optimal coagulant dosage in order to use it as on-line water quality monitor. Assuming that floc features at optimal dosages are unique, the overall objective was to find a way to use floc images. Besides, the equipment which is required for image analysis of flocs is not as expensive as typical water quality monitors.

Image Analysis in Coagulation – Update of literature review

The aggregation process of colloidal particles into larger flocs is a process that can proceed artificially or naturally influenced by various physical or chemical disturbance factors. Resulting flocs consist of a large amount of initial primary particles and have highly porous structures (Kim, 2001). Definition of flocs geometric parameters is a difficult task because of their amorphous, disordered nature. Since publishing the fundamental book of Mandelbrot (1983) main basis for floc structure analysis has become the Power Law. For example, the most widely used in the floc structure analysis – 2D fractal dimension, can be found from a power relationship between project area and characteristic length of flocs.

The first application of the Power Law for interpretation of alum floc structure had been done by Tambo (Tambo and Watanabe, 1979). Further, in his paper Li applied "fractal approach" to the application of image analysis of flocs generated during water and wastewater treatment (Li and Ganczarczyk, 1989). After publication of these two fundamental papers, "the fractal approach" in image analysis of different flocs had become a common technique (Kim, 2001; Maggi, 2006). At first papers, focus of researchers was on determination of floc settling velocity and calculation of respective floc density by direct observation of settling process in diluted model solutions (Milligan and Hill, 1998; Lee et al, 1996; Tambo and Watanabe, 1979). This approach gives sufficiently accurate results and can be operated *in-situ*. On the other hand, it is time consuming and complicated process. Moreover, it requires sensitive weighing system and therefore cannot be applied to industrial water monitoring systems.

At the same time, another sufficiently common approach for characterization of floc-structures is an image analysis based on the microphotography (Farrow and Warren, 1989). First images obtained during the coagulation process were taken by coupling the microscope with analog camera which gave a high enough magnification and good image details (Logan and Kilps, 1995; Gorczyca et al, 2003). This technique allowed to investigate the floc structure and morphology based on detailed analysis of microphotography (Cousin and Ganczarczyk, 1998). Besides, based on this technique correctness of fractal approach to the floc structure has been confirmed. However, as a previous approach, it was still highly time consuming technique because all operations on the image taking and analysis were performed manually. Moreover, this approach require sample extraction and/or special preparation in order to move it to microscope panel. Thus, this technique could not be used in industrial water monitoring systems.

The next step in image analysis was "digital approach" based on digital photo cameras and PCs (Logan, 1999; Chakraborti et al, 2000). Despite the relatively low resolution range of the digital cameras and low operating speed of PCs at that time this approach quickly became generally applicable. One reason was that coupling digital camera with PC considerably simplifies image taking and processing. Another reason is developing special technique of image taking by application of additional photo-chamber (Droppo, 2004). This technique allows to obtain images *in-situ* by cycle pumping suspension through the chamber. However, this could lead to flocs breakage because it is fragile nature (Jarvis et al, 2005). Nevertheless, the large number of papers based on this technique emerge last decade. As can be expected, with further development of digital cameras, PCs, software and new photo techniques "digital approach" for image taking and analysis could be even more applicable. Alternatively, there are few more new ways for *in-situ* image analysis that emerge last years: Particle Image Velocimetry (PIV) and Small Angle Laser Light Scattering (SALLS). These techniques are not quite new in general, but in application to image analysis they were not used so far. PIV analysis can be used to determine settling velocity as well as flocs shape and dimensions by analyzing their tracks via high speed image tracking system (Xiao et al, 2011). SALLS is used to determine floc concentrations and dimensions by analog or digital Fourier transformation of back scattered laser light (Kim et al, 2001). Both techniques could be used for calculation of fractal dimensions.

Methods, equipments and procedures

For jar tests 1 liter beakers equipped with Floaculator 2000 programmable mixer units (Kemira, Sweden) have been used. Mixing was performed in following modes: 1 min rapid mixing (400 RPM), 10 min slow mixing (30 RPM) and 20 min sedimentation without mixing. During the experiments 3 types of industrial coagulants were tested: ALS, PAX-18 and PIX-313 (Kemira, Sweden). To maintain stable pH value which equals 7 during the coagulation process 2M NaOH (VWR, Belgium) was added simultaneously with coagulant. During slow mixing stage accurate measurements of pH were performed in order to ensure that pH is stable and equals 7. After slow mixing stage mixer units were removed from glasses to avoid disturbance of flocs during sedimentation. Finally, after sedimentation stage approximately 200 ml of water samples were taken from 5 cm depth by peristaltic pump.

Listed in table 1 components in appropriate concentrations have been used to prepare model wastewater with reproducible parameters. Two types of hardness with corresponding alkalinities of initial wastewater have been selected in order to simulate typical Norwegian wastewaters and middle and western European wastewater types. Average initial wastewater parameters are listed in table 2.

Table 1 Model waste water composition (Ratnaweera, 1991)

No	**Component**	**Concentration level**		
		Low, mg/l	Medium, mg/l	High, mg/l
1	Dried Milk (Nestle, Norway)	150	300	600
2	Potato starch (Hoff, Norway)	30	60	120
3	Bentonite (Alfa Aesar, USA)	40	80	160
4	NaCl, soft/hard (Merck, Germany)	400/0	400/0	400/0
5	K_2HPO_4 (Merk, Germany)	25	50	100
6	NH_4Cl (Kebo, Germany)	50	100	200
7	Na-salt Humic acids (Merck, Germany)	2.5	5	10
8	$NaHCO_3$, soft/hard (Merck, Germany)	60/400	60/400	60/400
9	$CaCl_2$, soft/hard (Merck, Germany)	0/255	0/255	0/255

To obtain floc images during the coagulation process DSLR D600 camera (Nikon, Japan) with 105 mm Nikkor AF-S Micro 1:2.8 G ED lens (Nikkor, China) coupled with SpeedLite YN460 flash (Yongnuo, China) have been used. Following parameters were used during image taking process: ISO 800, F/32, 1/200. DigiCamControl 1.2.0 free software has been used for remote camera control to take images each 20 seconds during coagulation process. In addition, to obtain images modified mixing unit with attached 4 cm black plastic stripe was used, making a black background in images. Obtained images were processed in ImageJ free software to convert it to black and white, crop central 3000*3000 pixel area, enhance contrast and find GLCM texture parameters (Haralik et al, 1973) via “GLCM_TextureToo V 0.008” plugin (Cornish, 2007).

Each sample taken after sedimentation step was analyzed for total suspended solids, turbidity, ortho- and total phosphorous contents. The total amount of suspended solids was calculated as weight gain of the 1 hour dried at 105ºC Glass Fiber Filter (Whatman, UK) after filtration of a certain volume of water sample. Turbidity measurements were performed by 2100Q portable turbidimitre (Hach, USA). Concentration of ortho-phosphate ions in water samples were measured according to ISO 6678:2004(E) standard by using an UV5800PC spectrophotometer (Shanghai Bilon Instrument, China). Total phosphorous concentration were determined by using express Hach Lange photometric method with LCK 348 and/or LCK 349 standard test tubes and DR3900 spectrophotometer.

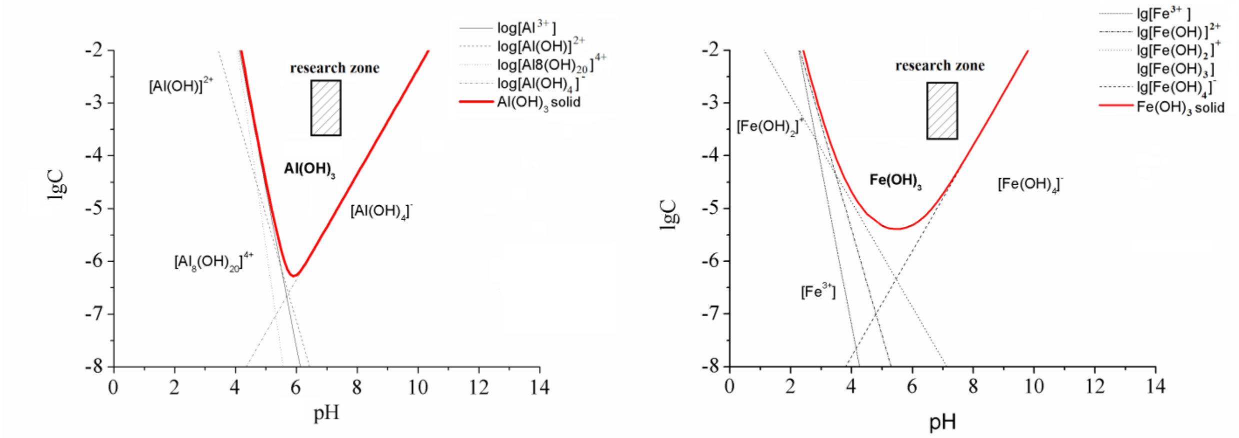
Table 2 Initial model wastewater parameters

Component	Concentration level		
	Low, (soft / hard)	Medium, (soft / hard)	High, (soft / hard)
pH	7.87±0.19 / 7.98±0.07	7.89±0.15 / 8.01±0.04	7.81±0.05 / 7.95±0.03
Turbidity, FTU	106±7 / 114±11	239±20 / 264±14	558±79/ 558±42
TSS, g/l	0.11±0.02 / 0.11±0.01	0.18±0.04 / 0.24±0.03	0.44±0.06 / 0.38±0.1
PO_4^{3-}, mg/l	4.72±0.3 / 5.01±0.1	9.96±0.99 / 10.17±0.7	19.7±0.1 / 17.63±0.1
Total P, mg/l	5.14±0.1 / 5.67±0.1	10.95±0.25 / 10.7±0.1	21.0±0.1 / 20.9±0.1

Experiments planning

During the experiments planning 2 types of initial model wastewaters (soft and hard) with 3 concentration levels of turbidity (low, medium, high) have been selected to simulate typical Norwegian and middle Europian wastewaters (table 1, 2). In addition, 3 types of coagulants (ALS, PAX-18, PIX-313) have been selected to reproduce the coagulation stage at typical wastewater treatment plants. pH value and concentration of coagulant during coagulation process have been selected by analyzing and reconstructing Amirtharajah coagulation mechanism diagrams to perform coagulation by sweep-floc mechanism (Amirtharajah and Kirk, 1982; Johnson and Amirtharajah, 1983). In preliminary research part 6 doses of coagulant were chosen with concentrations varied from 0.25 to 2.25 mmol/l to define the optimal coagulant doses for each water type. Graphical representation of coagulation initial conditions are shown in fig. 1 in appropriate solubility diagram for aluminum (a) and iron (b).

After defining optimal doses, 3 coagulant concentrations corresponding to near optimal, under- and over- dosages based on analysis of samples after coagulation process have been selected. Further, additional coagulation experiments with remote image taking system have been performed, while obtained images were processed in order to determine GLCM texture parameters.

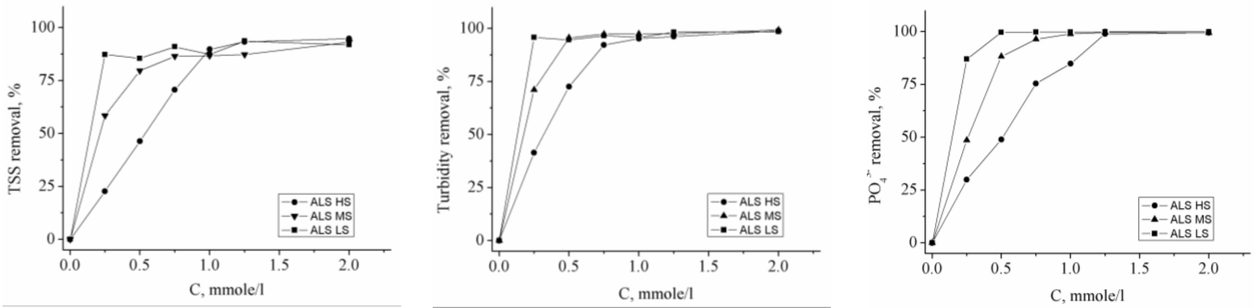


a) b)

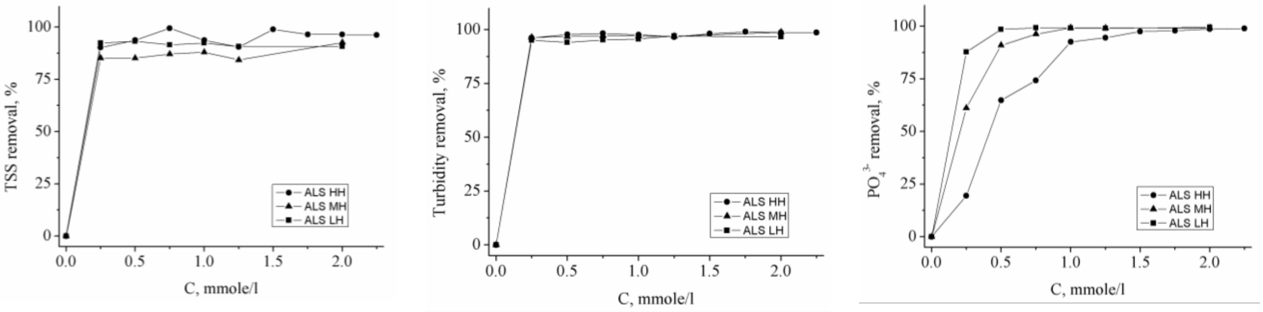
Figure 1 Coagulant solubility diagrams for Al(a) and Fe(III)(b) species (after Amirtharajah and Kirk, 1982; Johnson and Amirtharajah, 1983). Research zone highlighted by hatching.

Results and discussion

As was mentioned above, 2 types of model wastewater (soft and hard) with different concentration of components have been used for all experiments. Figure 2 shows the results of the jar tests with ALS coagulant while constant pH during coagulation.



a-1 b-1 c-1



a-2 b-2 c-2

Figure 2 ALS jar test results in TSS (a), turbidity (b) and ortho-phosphate (c) removal efficiency for soft (1) and hard (2) wastewater types with 3 contaminant concentrations: low, medium and high. pH=7.

Results show that soft wastewater types require higher coagulant addition for effective turbidity and TSS removal in comparison with hard wastewater types. For example, effective turbidity and TSS removal for soft wastewaters require addition of 0.50-1.00 mmol Al/l, while effective orthophosphate removal requires 0.50-1.25 mmol/l of Al for ALS coagulant. At the same conditions, hard wastewaters require 0.25-0.75 mmol/l Al for TSS and turbidity removal and 0.50-1.0 mmol/l Al for effective orthophosphate removal. Probably decreasing amount of coagulant to rich desired purification rate is influenced by the presence of Ca^{2+} ions, which enhance the coagulation process by double layer compression (O'melia, 1989). Almost the same trends have been observed for PAX-18 and PIX-313 coagulants. Thus, based on obtained results 1 mmol/l equivalent dosage of Al has been selected for further investigations as a near optimal dosage, 0.25 mmol/l Al – under- dosage and 2 mmol/l Al – over- dosage.

Naturally, during coagulation process particles and aggregates have tend to growth until the system will reach a steady state point. At equilibrium resulting flocs are stable because of equal rate of aggregation and disaggregation processes. During steady state floc structure could be reproducibly defined, as it is not vary significantly. Thus, to define a steady state time, images of coagulation process were collected each 20 seconds during a slow mixing period. Next, resulting set of images was processed and GLCM texture parameters have been obtained. Further, the statistical relationships between GLCM texture parameters and time have been performed using Principal Component Analysis. Fig. 3 shows the scores response of the first principal component (PC1) of GLCM image texture features as a function of time during coagulation process (slow mixing period).

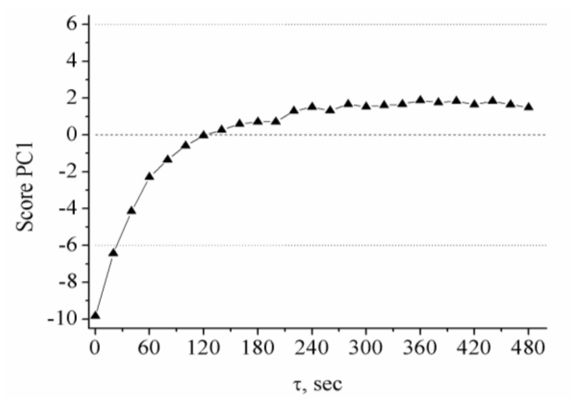
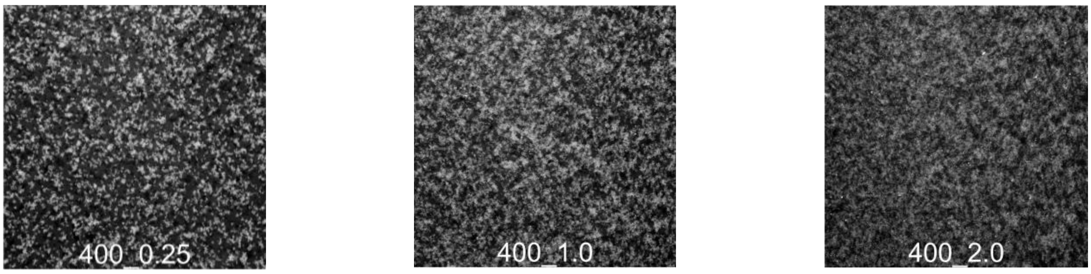


Figure 3 Response of PC1 GLCM texture parameters as a function of slow mixing time: ALS; 0.25 mmol/l; Low Soft wastewater type.

Analysis of fig. 3 shows that at first images structures are tend to change until the steady state region is reached in 250-400 seconds (in overall for all wastewater types and coagulants). Further, it tends to show the stability of GLCM texture parameters of images, thus, it proves the certain level stability of flocs structures after some time of slow mixing. This time interval have been used in further research for various coagulant dosages. It should be noted, that obtained results are in a good agreement with the previous studies (Sivchenko et al, 2013), where the results were obtained by analyzing physical dimensions of particles and using Angle Measure Technique (AMT) for images during coagulation process. Hence, GLCM texture analysis of floc images obtained during coagulation process can be used as a tool for prediction of image texture structure. Assuming that floc features at optimal dosages are unique, this technique potentially could be used to predict the optimal coagulant dosages.

To verify this hypothesis, further investigations of GLCM texture features for optimal coagulant dosages and different wastewater types were conducted. Resulting images of flocs for ALS coagulant and medium soft model wastewater type at the same coagulation time are shown in fig. 4.

Even without image analysis it could be concluded that there is a strict difference between obtained images. In order to find statistical relationships between GLCM texture parameters and coagulant doses PCA analysis for all wastewater types has been performed.



a) b) c)

Figure 4 Floc images obtained by addition of 0.25 (a), 1.00 (b) and 2.00 (c) mmol/l of Al: ALS; 400 seconds after slow mixing started; medium soft wastewater type.

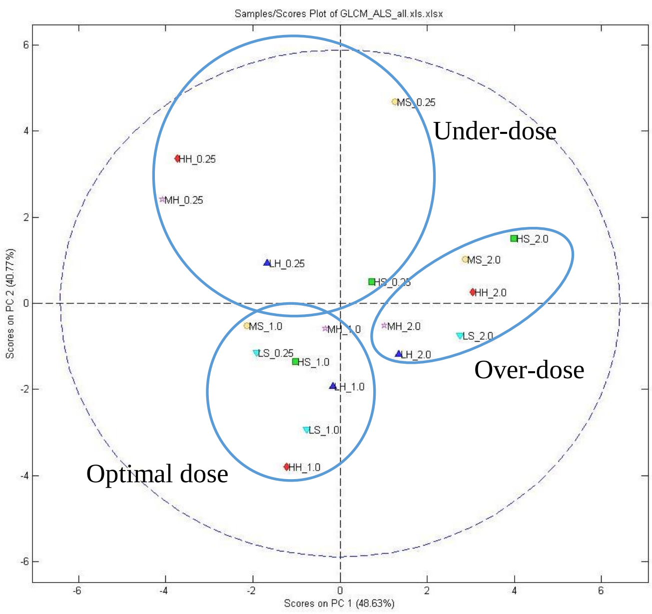


Figure 5 Scores on PC1 and PC2 in PCA of GLCM texture parameters with different wastewater types

Fig. 5 shows that the responses of PC1 versus PC2 of GLCM texture parameters are located at 4 different zones of PCA scores graph. Most part of under- dosages are located at negative PC1 and positive PC2 zone, while remaining part that locates at other regions is more related to optimal dosage than to the under- dosage according to treatment efficiencies results. Meanwhile, all optimal dosages are located at negative PC1 and negative PC2 zones. At the same time over- dosages are located in positive PC1 and positive and negative PC2 zones. Thus, as different dosages are located in different regions of PCA graph, they can be principally separated to 3 independent groups that corresponds to different coagulant dosages based on PCA scores. In particularly, this allow to predict coagulant dosage based on analysis of images obtained directly during coagulation process.

Conclusions

GLCM texture analysis is a promising technique for water and wastewater quality monitoring. Based on obtained results it could be concluded that there is a strict correlation between image GLCM texture features and physical parameters of flocs. Moreover, using PCR or PLSR for GLCM texture parameters optimal dosages of coagulant could be predicted.

Recommendation for further research

As observed correlations are not as clear for PAX-18 and PIX-313 coagulants more detailed investigation with more than 3 dosages of coagulant should be performed. Next, investigation on real wastewater should be performed in order to identify potential application of this technique to wastewater quality monitoring.

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