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Novel inorganic-organic composite coagulants based on aluminium

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ABSTRACT

In this study, the preparation of a novel coagulant is examined, by introducing into PACl a cationic polyelectrolyte (CPE). For this purpose, poly-diallyldimethylammonium chloride (p-DADMAC) was used, a commonly applied cationic polyelectrolyte in water and wastewater treatment. Several possible derivatives of the composite coagulant were prepared, based on different CPE content (Al/CPE ratio, w/w). The products were characterized by means of polymerization degree (aluminium species distribution), chemical bonding (FT-IR spectroscopy), conductivity, turbidity and pH, in order to examine the impact of CPE addition in the properties of PACl. Moreover, their coagulation performance was evaluated for the treatment of contaminated tap water and compared to the respective performance of the combined application of PACl and CPE but as separated reagents. Finally, the kinetics of coagulation-flocculation was studied by aims of a Photometric Dispersion Analyzer (PDA). From the results obtained it was revealed that despite the deterioration of the charge neutralization capability when the two reagents are combined into one, the composite coagulants are more efficient in water treatment due to more effective particles aggregation. The most important advantages of the new coagulants are the significant reduction of CPE dosage (or content) and the lower residual aluminium concentration. Specific attention has to be given to the CPE content, in order to produce an improved coagulant which can achieve efficient charge neutralization combined with enhanced particles aggregation, leading to more efficient and cost-effective treatment.

Keywords: Polyaluminium chloride; Composite coagulants; Cationic polyelectrolyte; p-DADMAC; Preparation; Characterization; Application

1. Introduction

Significant progress has been achieved during the last two decades in the coagulation–flocculation field. A new category of coagulation reagents was developed, the so-called pre-polymerized coagulants, which result after the controlled partial polymerization of several simple metal salts (or conventional coagulants, i.e. $AlCl_3$, $FeCl_3$, $FeSO_4$, etc.). The properties of the new reagents were intensively examined and have proved to be more efficient in lower dosages, in wider pH, temperature and colloids concentration ranges, than the conventional simpler ones, leading to cost and operative more effective treatment [1,2]. However, despite their improved properties, the efficiency of the pre-polymerized coagulants still remains inferior when compared to the performance of organic polyelectrolytes, e.g. when used in water treatment [3].

The main reason for the higher efficiency of organic polymers is their higher molecular weight (MW), which implies better flocculation properties. Thus, the increase of molecular weight and size of the

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pre-polymerized coagulants ingredients is thought to be the way for further improvement. This increase can be achieved through the combination of a pre-polymerized coagulant and a suitable additive (inorganic or organic) into one reagent. The general concept followed is, the introduction of various additives in the structure of a pre-polymerized coagulant, in order to produce a homogenous, stable product with higher MW and improved coagulation-flocculation performance, than the initial reagent. The challenge to confront is the desirable combination of higher efficiency and cost-effectiveness, which are the basic prerequisites for the development of new products.

Regarding the organic additives, the combination of an inorganic salt and a suitable organic polymer (polyelectrolyte) into one reagent is thought to improve, apart from the treatment efficiency of the inorganic salt, the overall cost-effectiveness. No addition of a flocculant aid will be needed any more, whereas the introduction of the polyelectrolyte into the structure of the metal salt will also significantly reduce the eventual residual toxicity due to remaining unreacted monomers of the polymer [4].

Synthetic polyelectrolytes have been utilized in coagulation/flocculation process for water purification for more than four decades, whereas natural polymers are known as flocculants since ancient times. Their principal uses are as primary coagulants (cationic polyelectrolytes) as well as in the more traditional flocculation step of binding already formed small flocs into larger aggregates (flocculant aids, anionic and nonionic polyelectrolytes) [5]. An important group of synthetic cationic polyelectrolytes are those derived from diallyldimethylammonium chloride (DADMAC) polymerization. DADMAC is synthesized from allyl chloride and dimethylamine, as shown in Fig. 1. The homopolymer derived from DADMAC polymerization is referred as p-DADMAC and can have a MW up to 2–3 × 10^6 Da. p-DADMAC products are available as viscous aqueous solutions at concentrations up to 40% w/w and are used extensively in water or wastewater treatment as primary coagulants [5,6]. Usually they are used in conjunction with inorganic metal salts, thus achieving a reduction of the inorganic coagulant dosage up to 80%.

In this study, several composite coagulation reagents derived from the combination of the cationic polyelectrolyte p-DADMAC and the pre-polymerized coagulant polyaluminium chloride (PACl) were prepared. The value of OH/Al molar ratio was chosen to be 2, because pre-polymerized coagulants with this OH/Al value proved to be more efficient in water treatment [7]. The p-DADMAC content, expressed as Al/p-DADMAC w/w ratio (mg/L/mg/L, denoted as Al/CPE) ranged from 2.5 to 10. The impact of p-DADMAC addition in the initial aluminium species distribution (in PACl) was investigated using the spectrophotometric ferron technique. An FT-IR study was also conducted, in order to observe possible alterations in the chemical bonds after p-DADMAC addition. These alterations could be indicative of the formation of new species, arising from the interaction of p-DADMAC with Al species, whereas the absence of such alterations would indicate that the new coagulants are simple mixtures of the two reagents. Furthermore, their coagulation efficiency was evaluated for the treatment of simulated surface water and compared to the respective coagulation performance of the combined addition of p-DADMAC and PACl, but as separated reagents. It should be mentioned that the addition of a cationic polyelectrolyte in a PACl solution is expected to increase, apart from the molecular weight, the charge density of the composite coagulant, thus enhancing the charge neutralization capability and resulting in a more effective coagulation (destabilization of colloids) as well.

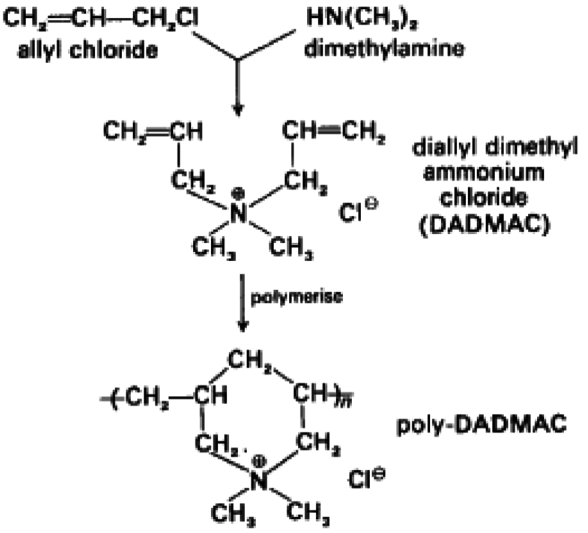

Fig. 1. Synthesis route of p-DADMAC [6].

2. Materials and methods

2.1. Synthesis of composite coagulation reagents

All used chemical reagents were analytically pure chemicals. Deionized water with conductivity lower than 0.5 μS/cm was used to prepare all the solutions, except of the solutions used for the synthesis of the coagulants. In this case, deionized water made carbonate free by boiling, was used.

Table 1
Properties of the model sample to be treated and jar-test conditions

Model sample properties			Jar-test experimental conditions			
Turbidity	Absorbance at 254 nm	pH	Rapid mixing stage		Slow mixing stage	
			Mixing rate	Duration	Mixing rate	Duration
16.5 NT U	0.125	7.65	160 rpm	120 s	45 rpm	10 min

First, intermediate PACl solutions were prepared according to the following procedure: a predetermined amount of 0.5 M $AlCl_3.6H_2O$ was placed in a baker on a magnetic stirrer (Al solution). Appropriate amount of 0.5 M NaOH in order to achieve the desired OH/Al = 2 molar ratio was slowly added (with the use of a peristaltic pump) in the aluminium solution under intensive stirring. The base addition rate was 0.1 ml/min and the stirring speed 800 rpm. Then, the appropriate amount of p-DADMAC was weighted (according to the desired Al/CPE ratio), diluted to 10 ml with deionized water and slowly added (addition rate 0.2 ml/min) under magnetic stirring in the PACl solution.

After the preparation, the composite coagulants and PACl were diluted to 0.1 M Al. The coagulants are referred in the text as follows: composite coagulant with e.g. Al/CPE = 5 and OH/Al = 2 as PACPE 2/5 and PACl with OH/Al = 2 as PACl 2.

2.2. *Characterization methods*

The pH was measured by using a Metrohm Herisau pH-Meter, conductivity by using a Crison CM 35 Conductivity Meter and the turbidity measurements were performed on a HACH RATIO/XR Turbidimeter.

2.2.1. *Aluminium species distribution*

Aluminium species distribution was determined with the application of Al-ferron timed spectrophotometric method, which is based on the different reaction time of aluminium species with ferron reagent (8-hydroxy-7-iodoquinoline-5-sulphonic acid) to form water soluble complexes in pH 5–5.2. These complexes absorb light with a maximum at 370 nm, hence absorbance measurements at this wavelength allow the calculation of the different species of aluminium. A UV–Vis spectrophotometer (Shimadzu) was used for this purpose. The exact procedure was a modification of the method of Parker and Bertsch [8], developed by Zhou et al. [9].

2.2.2. *FT-infrared spectroscopy (FTIR)*

Specific amounts of samples were placed in glass bakers and kept in an oven at about 40 °C for several days, in order to obtain dried powders. FTIR spectra were recorded in the range of 4,000–400 cm^{-1} using a Perkin Elmer Spectrophotometer (Perkin Elmer, Spectrum One). The pellet was prepared by mixing 1 mg of the aforementioned powders with 200 mg KBr.

2.3. *Evaluation of coagulation performance*

For the determination of coagulation efficiency of the prepared coagulants a jar-test apparatus (Aqualytic) with six paddles was used. The treated sample (1 L) was made of tap water, clay (kaolin) suspension (commercially available) and humic acid (Aldrich). The initial concentration of clay suspended particles was 10 mg/L and that of humic acid 5 mg/L. Table 1 displays the properties of the model sample as well as the jar-test experimental conditions. One min after the addition of the coagulant (i.e. during the rapid mixing stage) 30 mL sample was withdrawn for z-potential measurements. No flocculant aid was used and at the end of these experiments about 50 ml of sample was withdrawn 5 cm below the liquid surface for further analytical determinations. The concentrations of coagulants are expressed as mg (Al + p-DADMAC)/L in both cases, i.e. when PACl and p-DADMAC are applied together but as different reagents or when the composite coagulants are applied. In the first case, the calculated amounts of PACl and p-DADMAC are introduced at the same time in the treated sample. Settling time was 40 min.

The zeta-potential was measured by using a Laser Zee Meter 501, absorbance at 254 nm as a convenient indicator of organic matter presence was measured with a Schimadzu UV/Vis spectrophotometer using a 1 cm path length quartz cuvette. Residual aluminum concentration was determined with eriochrome cyanine R method [10].

2.4. *Study of coagulation kinetics*

The extent of aggregation was also examined and the flocculation dynamics was accomplished by using

Table 2
Physicochemical properties and Al species distribution in the prepared coagulants

Coagulant	Physicochemical properties			Al species distribution		
	Turbidity (NTU)	Conductivity (mS/cm)	pH	Al_a	Al_b	Al_c
PACl 2	2	25.8	4.39	16	69	15
PACPE 2/10	2.5	26.2	4.34	15	72	13
PACPE 2/5	2.8	26.5	4.37	12	75	13
PACPE 2/2.5	4	26.7	4.35	11	70	19

a continuous flow optical flocculation monitor (PDA 2000, Rank Brothers, UK). The test suspension of 1.5 L tap water, containing 10 mg/L clay and 5 mg/L humic acid, was placed in a 2 L beaker and stirred with the paddle of a jar-test apparatus. The suspension flows through the measuring transparent plastic cell (with 3 mm diameter), where it was illuminated by a narrow light beam (of 850 nm wavelength), with the help of a peristaltic pump. The pump was placed after the PDA apparatus for preventing the eventual floc breakage caused by the mechanical forces of the pump. The applied flow rate was 30 ml/min in order to have laminar conditions in the sampling tube, hence avoiding flocs breakage. The rest experimental conditions were the same with the jar-tests experiments, except that slow mixing lasts until the end of the experiments. Concentration of coagulants was 2 mg/L and pH 7.65 (initial).

3. Results and discussion

3.1. Characterization of the prepared coagulants

Table 2 lists the properties of PACl and composite coagulants prepared in the laboratory. Regarding the physicochemical properties, it can be seen that the addition of p-DADMAC in PACl results in an increase of PACl turbidity, indicating a possible increase of the coagulant components size. By increasing CPE content (i.e. decreasing Al/CPE ratio) turbidity increases, and the highest turbidity value exhibits PACPE 2/2.5. It should be mentioned that despite the increase of turbidity, no solids formation was observed in all composite coagulants, and it can be assumed that the final product was a homogenous liquid. pH value seems to be unaffected by the addition of p-DADMAC, whereas a slightly increase of conductivity can be observed by increasing the amount of p-DADMAC in the composite coagulants.

Aluminium species distribution strongly correlates with the coagulation performance of an Al coagulant. With the use of ferron reagent, the Al species can be categorized into three main fractions; Al_a which corresponds to monomeric Al species, Al_b which corresponds to medium sized Al polymers and Al_c which corresponds to large Al polymeric species. The most important fraction is thought to be Al_b, as the most effective Al specie (i.e. Al_{13}) regarding coagulation efficiency is included in this fraction. From Table 1 it can be seen that the addition of p-DADMAC results in a slightly increase of Al_b fraction in the composite coagulants, and a slightly decrease of Al_a and Al_c fractions. The decrease of Al_a is more intense than the respective decrease of Al_c. The introduction of the polyelectrolyte increases the effective Al species fraction, possibly having a positive effect in the coagulation performance of the composite coagulants.

In order to extract a more completed picture about the composition of the prepared coagulants, the possible chemical bonds present in the prepared reagents were examined. The IR spectra of composite coagulants in the region of 4,000–400 cm^{-1} were compared with the respective IR spectra of PACl 2 and p-DADMAC, all dried in an oven under relatively low temperature (<40 °C).

Fig. 2 displays the aforementioned spectra. A detailed analysis of PACl spectra can be found in

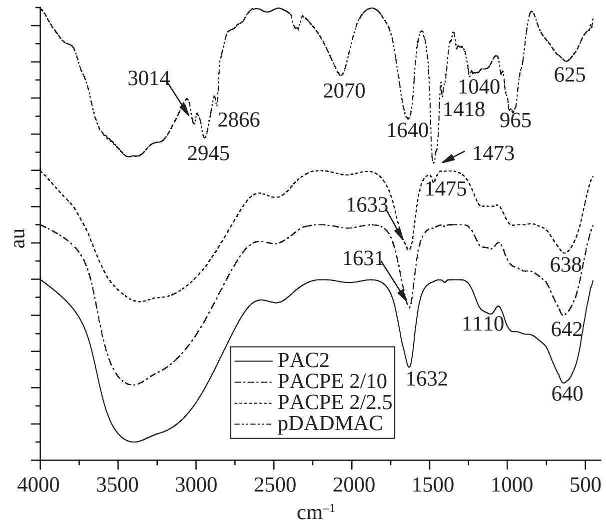

Fig. 2. FT-IR spectra of examined coagulants and p-DADMAC.

Table 3
Analysis of FT-IR spectra of a polyaluminium chloride solution (with OH/Al = 2) [11]

Wavelength (cm^{-1})	Band assigned to
3400	Hydroxyl –OH stretching vibration
1632	H_2O molecules
1160	bending vibrations of Al-OH_2
1110	oxo groups or oxo bridges
980	bending vibrations of Al-OH_2
640	symmetric stretching mode of Al-O bond of the central AlO_4^- in Al_{13} molecule

[11]. Briefly, the bands expected to appear in the IR spectra of PACl are those associated with OH vibrations of water or bridging hydroxyls and with Al-O bond vibrations. Table 3 summarizes the most important bands of a polyaluminium chloride sample. Regarding p-DADMAC, the most important bands are the following: at 3,014 cm^{-1} due to C-H stretching vibrations, at 2,945 and 2,866 cm^{-1} due to asymmetric and symmetric –CH_2 vibrations, at 2,070 and around 1,000 cm^{-1} appear the stretching vibrations of –CN, the band at 1640 is assigned to stretching vibrations of C = C, at 1,473 cm^{-1} assigned to -CH_3 vibrations and at around 1,400 cm^{-1} to stretching vibrations of N-H [12,13]. For both samples, the bands at the region 3,400–3,500 cm^{-1} are associated with water molecules vibrations.

From Fig. 2 it can be seen that the addition of p-DADMAC does not alter significantly the bands of the initial PACl coagulant. The most noticeable differences can be observed in the region of 1,000–1,500 cm^{-1}. Specifically, by increasing the p-DADMAC content a new band appears at the wavelength of 1,475 cm^{-1} and at the same time the band at 1,110 cm^{-1} disappears. The new band appearing at 1,475 cm^{-1} is due to the high intensity band of –CH_3 in p-DADMAC, and does not indicate the formation of new species in the composite coagulants. The disappearance of the band at 1,100 cm^{-1} can be associated with the increase of –CN content and its corresponding band at around 1,000 cm^{-1}, which results in overlapping of these two bands. Another alteration is the increase of the band width at around 1,630 cm^{-1}, probably resulting from the overlapping of C = C band (1,640 cm^{-1}) of p-DADMAC and H_2O band ($\sim$1,630 cm^{-1}) of PACl.

The majority of Al species in PACl are positively charged, so an electrostatic interaction between these species and positively charged p-DADMAC can not take place. One possible interaction which could lead to new species formation is with the only negatively charged Al specie, i.e. $Al(OH)_4^-$. However, considering that $Al(OH)_4^-$ is a hydrolysis product of Al dissolution and exists at pH > 7.5, and that PACl sample is a neutralized Al solution (most of Al anion consumed for Al_{13} formation [7,11]) and has a relatively low pH, i.e. pH < 4.5, the possibility of this kind of interactions is relatively low. Moreover, from the IR spectra evidence regarding new bonds formation was not revealed, and it can be initially assumed that no new chemical species are formed when p-DADMAC is added to pre-neutralized Al solution, at least not in a noticeable degree. It is more likely that the composite coagulants comprise of a simple mixture of Al species and p-DADMAC. However, some interaction should take place, as the Al species distribution in PACl is altered to a certain extent (slight increase of Al_b).

3.2. Evaluation of coagulation performance

Before the comparative coagulation experiments, preliminary jar tests were conducted using PACl and p-DADMAC (or CPE) in conjunction but as separated reagents, in order to determine the optimum ratio of PACl and p-DADMAC (w/w) achieving the best performance in treating the model sample. Several ratios ranging from 1:1 to 20:1 (Al/CPE, w/w) were examined, and from these experiments it was concluded that the optimum Al/CPE ratio was 2.5:1 (data not shown).

Fig. 3a displays the turbidity reduction relative to coagulants concentration. For very low dosage (i.e. 1 mg/L) of coagulants the application of PACl and p-DADMAC seems to be more effective in turbidity removal. However, the efficiency can not be assigned as satisfactory, as the final turbidity value is far greater than the limit of 1 NTU for drinking water. By increasing the coagulants concentration, the composite coagulants exhibit better performance. For 3 mg/L and higher dosages, turbidity values remain under or close to 1 NTU for the composite coagulants, whereas with PACl and CPE for dosages greater than 5 mg/L turbidity value of the treated sample increases, indicating the negative overdosing effect. Regarding the composite coagulants, better performance exhibit those with Al/CPE ratio 5 and 10. The performance of PACPE with Al/CPE 2.5 is similar to the respective performance of PACl and CPE (as separated reagents).

Regarding organic matter removal, measured as UV absorbance at 254 nm reduction (Fig. 3b), similar behavior of the tested coagulants can be observed. Particularly, again the combined application of the separated reagents PACl and CPE is more efficient in very low dosages (i.e. 1 mg/L), but with increasing coagulants dosages, the composite coagulants exhibit greater removal efficiencies. After the dosage of 2 mg/L, no

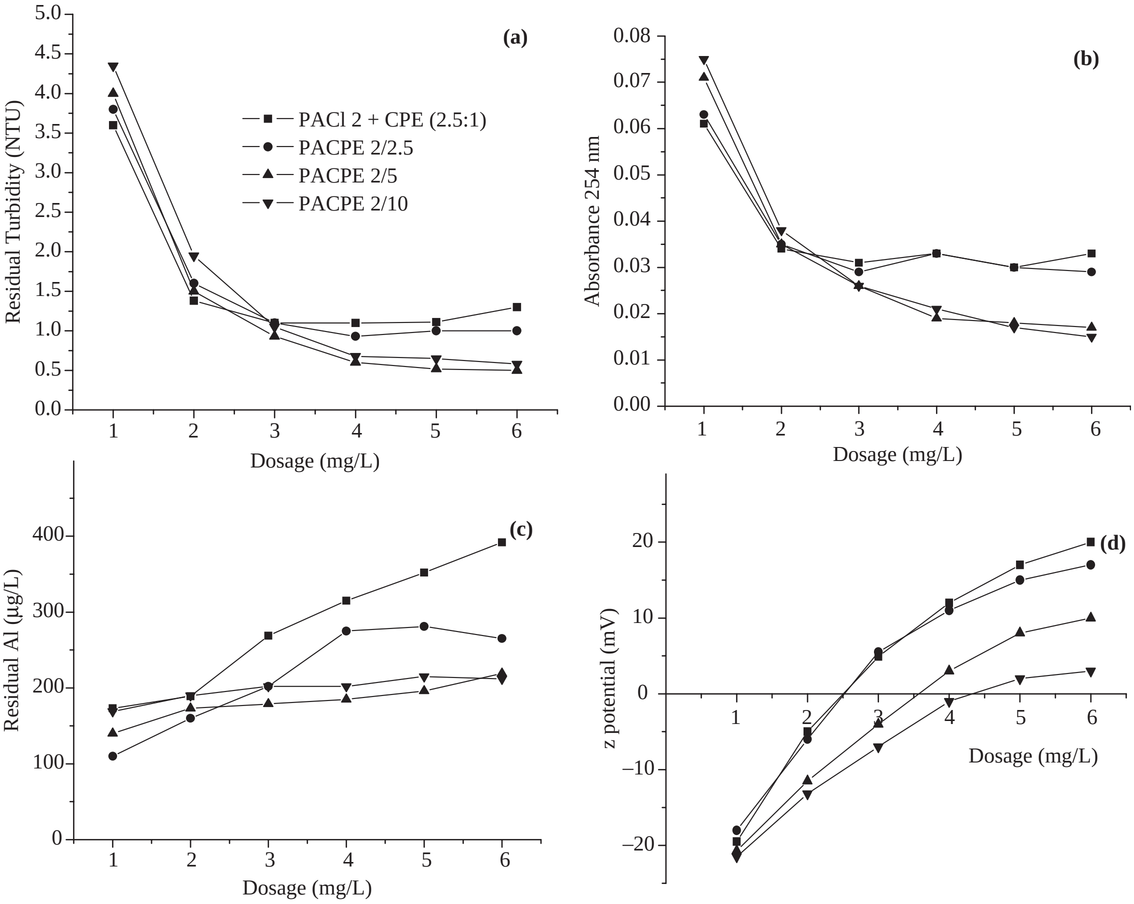

Fig. 3. Coagulation experiments results. (a) residual turbidity (initial turbidity 16.5 NTU), (b) UV absorbance at 254 nm (initial absorbance 0.125), (c) residual aluminum concentration, (d) z-potential measurements.

significant changes are observed in abs 254 nm in the case of PACl and CPE and PACPE 2/2.5, and again the coagulation behavior is similar when PACl and CPE are applied as separated reagents or as one composite reagent with the same Al/CPE = 2.5 ratio.

A very important parameter, which designates at a certain degree the possibility of the application of a coagulant in water treatment, is the residual aluminium concentration that remains in the treated sample. From Fig. 3c it can be observed that in the case of composite coagulants with Al/CPE 5 and 10 the residual Al concentration remains under or close to 200 μg/L, which is the EU maximum permissible concentration limit for drinking water (EU Dir, [14]). The same pattern can be observed almost in all cases, i.e. when the coagulants concentration increases, the Al concentration that remains in the treated sample also increases. This increase is more intense when PACl and p-DADMAC are applied as separated reagents, and in this case the highest residual Al concentrations can be observed. The differences with the composite coagulants are quite large, even when compared to PACPE with the same Al/CPE 2.5, indicating the significance of the Al species and p-DADMAC molecules complexes formation. The incorporation of p-DADMAC in the structure of PACl increases the size of the initial Al species, which probably form larger flocs and are separated more efficient through gravity settling, thus leaving smaller amounts of Al species in the treated sample. This advantage is very important, and together with the aforementioned treatment results, the superiority of the composite coagulants is more than clear.

Fig. 3d represents the alteration of the colloids z-potential with increasing coagulants concentration. Z-potential measurements offer an indirect estimation of the impact that the coagulants exhibit in the particulates charge, and moreover they allow comparisons to be made between different coagulants. With increasing coagulants dosages, z-potential values increase for all cases and convert from negative to positive values. This increase is highly depended on p-DADMAC amount in the composite coagulants, i.e. by increasing p-DADMAC amount (or decreasing Al/CPE ratio) z-values increase more intense. It can be concluded that the introduction of a cationic polyelectrolyte significantly increases the charge density of PACl, resulting in a more effective charge neutralization capability. Therefore, it is reasonable to expect that the composite coagulants with the highest polyelectrolyte content should be the most efficient reagents regarding coagulation. However, this assumption was not supported from the coagulation experiments results. Specifically, it was observed that the composite coagulant with the highest p-DADMAC content (i.e. Al/CPE = 2.5) was not the most efficient coagulant, indicating the significance of flocculation in the whole process. The appropriate amount of p-DADMAC should be added into PACl, in order to produce Al-pDADMAC species of the optimum size, which in success will achieve the best treatment efficiency. From the results obtained, the optimum Al/CPE ratio should be 5 or 10. Moreover, if the CPE content is too high (i.e. 2.5), the z-potential values increase in a high extent, especially for dosages higher than 4 mg/L. As a result, re-stabilization of the colloids occurs (overdosing effect), explaining the worsening or not improving of the performance of PACPE with OH/Al 2.5 for dosages greater than 4 mg/L.

Comparing the z-potential values variation when PACl and CPE are applied as separated reagents or as a unique coagulant (Al/CPE = 2.5), it can be seen that the impact in the particulates charge is similar for both cases up to the concentration of 3 mg/L. For greater coagulant dosages, the application of PACl and p-DADMAC as separated reagents exhibits greater impact in particulates charge, and this difference is increasing with the further increase of coagulants concentration. The slight deterioration of charge neutralization capability in the composite coagulants with increasing polyelectrolyte content can be due to oversaturation effects, which lead to polymer chain compression and loss of a certain percent of charged sites, which are not available any more for interaction with colloids negatively charged sites. However, this effect is noticeable only when relatively high dosages of coagulants are applied. Another possible oversaturation effect could be the decrease of the distance between Al species and p-DADMAC molecules, thus leading to interactions between specific sites of Al species (e.g. bridging hydroxyls) and p-DADMAC (e.g. $=N^+=$). From this point of view, the formation of new species in the composite coagulants can not be excluded, despite the fact that such evidences were not observed with FT-IR investigation. However, it is more likely that this alteration takes place at relatively small extent, as the modification of Al species, as revealed through the ferron technique, was very small.

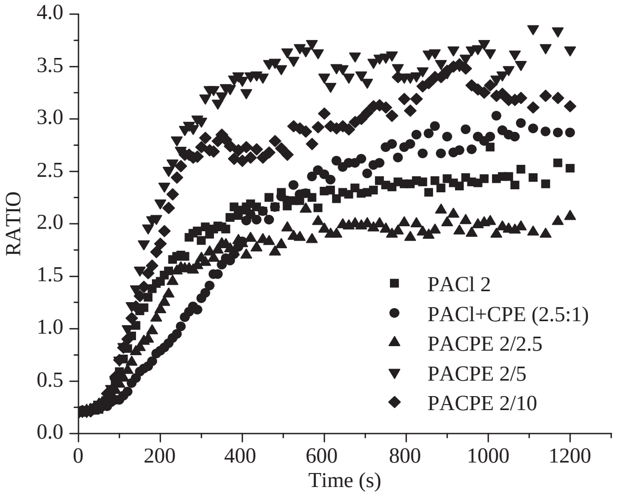

Fig. 4. Study of the coagulation kinetics for all prepared coagulants.

3.3. Study of coagulation kinetics

The PDA apparatus measures the average transmitted light intensity (dc value) and the root mean square (rms value) of the fluctuating component. The ratio (rms/dc), or Flocculation Index (FI) provides a sensitive measure for the aggregation of particles. The FI value is strongly correlated with the respective floc size and always increases as flocs grow larger, providing a useful (although relative) indication of floc growth, eventually breakage and re-growth, which allows comparisons to be made between the different coagulants and under different shear conditions and coagulant concentrations [15].

The overall process of treatment with coagulation reagents can be divided into two procedures, taking place subsequently, i.e. the coagulation (destabilization of the colloids), which takes place shortly and immediately after the addition of coagulant, and the flocculation, where the previously destabilized particles aggregate to form larger flocs (usually after the appropriate addition of organic polyelectrolytes). During the initial coagulation stage (rapid mixing period), the FI or Ratio (R) remains in low values (lag phase) and begins to increase suddenly, when the subsequent flocculation stage (slow mixing period) takes place. As flocculation proceeds, the Ratio increases up to a maximum value and finally, equilibrium between the formation and the breakage of flocs occurs, where the Ratio values remain relatively constant. The final plateau of R-values shows the obtained degree of particle aggregation [7,15].

Fig. 4 illustrates the Ratio (or FI) values variation against time (s) for all prepared coagulants. The highest ratio values are attributed to the composite coagulants with Al/CPE 5 and 10, indicating greater floc size for these coagulants, and between these two reagents

PACPE 2/5 seems to generate bigger flocs. These observations confirm the assumptions made in chapter 3.2. Moreover, when PACl and p-DADMAC are applied as separated reagents, the lag phase (i.e. the time period from the beginning until the maximum Ratio values are attained) is significantly increased, compared to the composite coagulants. The shorter duration of lag phase is desirable in water treatment due to energy and time savings, and therefore the composite coagulants exhibit an additional advantage, the quicker flocs formation. Finally, from Fig. 2 it can be seen that the introduction of p-DADMAC in PACl (composite coagulants) leads to bigger flocs formation than PACl alone only when Al/CPE ratio is 5 or 10, the optimum values.

4. Conclusions

The combination of a cationic polyelectrolyte (p-DADMAC) and an inorganic pre-polymerized coagulant (PACl) into one composite reagent (PACPE) exhibits several advantages against the application of those as separated reagents. Better treatment efficiency was achieved with significantly lower polyelectrolyte amount (i.e. two or four times lower: Al/CPE 5 or 10 compared to Al/CPE 2.5). In particular, it was found that the most important advantage of the composite coagulants is the significantly lower residual Al concentration that remains in the treated sample. It seems that the deterioration of the charge neutralization capability when PACl and CPE are combined to one reagent is compensated by the formation of flocs of greater size, as shown by PDA experiments, resulting in a more effective treatment performance. A positive effect may also exhibit the alteration in a certain extent of aluminium species distribution, i.e. slight increase of Al_b content in the composite coagulants. This alteration could be due to new species formation, despite the fact that such evidence was not obtained through FT-IR analysis, as explained in the text. Concluding, the promising results obtained can serve as basis for further research regarding the development of the inorganic-organic composite coagulants.

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