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REMOVAL OF ARSENATES FROM CONTAMINATED WATER BY COAGULATION–DIRECT FILTRATION

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ABSTRACT

The present study has been directed towards the removal of arsenic, applying a modification of conventional coagulation/-flocculation process. The modifications refer to the introduction of "pipe flocculation" process in the first stage of the technique, whereas the second step has been performed by direct filtration with sand filters, instead of separation by sedimentation. Alum or ferric chloride was the coagulant agent used, enhanced in certain cases by the presence of cationic or anionic polyelectrolytes (organic polymers). The efficiency of coagulant/polymer addition was investigated, using different ratios and for different initial As(V) concentrations. The effect of linear velocity during sand filtration was also examined. In general, both the coagulants were found to be efficient regarding arsenic removal and in both cases the use of coagulant aids increased the overall efficiency of the method—reaching in some cases arsenic removals up to 99%.

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Key Words: Arsenate; Coagulation; Direct filtration; Water treatment

INTRODUCTION

Arsenic is widely known for its high toxicity to humans. Chronic arsenic intake of more than 100 μg/L can cause skin and lung cancer, as well as other diseases.[1–3] Thus, the European Commission reduced the limit of arsenic in drinking water from 50 to 10 μg/L.[4] This revision would certainly have direct consequences regarding the maximum concentration of arsenic allowed in industrial wastewaters, where usual arsenic concentrations fall in the range 0.5–2 mg/L.[5]

Arsenic contamination of wastewaters is often caused by agricultural activities, where arsenic is used for the production of certain insecticides and herbicides. Areas that are close to mining activities or smelters have been also found to contain elevated arsenic concentrations.[6] These are considered as anthropogenic sources of arsenic contamination and treatment of these wastewaters is necessary, in order to reduce the arsenic content, in such a way that it would not cause problems to natural waters when disposed.

Inorganic arsenic may be present in two oxidation states, as arsenate [$H_2AsO_4^-$] or arsenite [H_3AsO_3]. Dominant arsenic species are a function of pH and redox potential conditions. Arsenate is the thermodynamically stable form of inorganic species and it generally predominates in surface waters. Arsenite is favored under reducing conditions, as for example in anaerobic groundwaters. Organic arsenic species may also occur in natural waters as a result of the use of organo-arsenical pesticides, as well as through the biomethylation mechanisms of microorganisms. It is well established that the toxicity of arsenic depends on its chemical form. Arsenite, the trivalent form is far more toxic in biological systems than arsenate. The toxicity of organo-arsenicals is generally lower than that of inorganic arsenic species.[7]

Several methods have been developed for the removal of arsenic from contaminated water sources or from industrial wastewaters. The most important among them are: (a) coagulation/filtration, (b) adsorption on activated alumna, (c) lime softening, (d) ion-exchange, (e) reverse osmosis, (f) adsorbing colloid flotation, and (g) adsorptive filtration.[8–14] These methods are mainly effective for the removal of pentavalent arsenic. Therefore, a pre-oxidation step is usually required, in order to remove effectively the trivalent arsenic content.[8] In this paper, the modified method of coagulation/direct filtration was selected for further research, as it is a relatively common method in wastewater treatment, presenting several advantages; it uses relatively low costs, simple chemical reagents; it has low capital cost, it is a compact method, and it does not need monitoring of the breakthrough point, as in column-adsorption processes. On the other hand, this method produces toxic sludge, which has to be further treated and disposed safely.

The treatment of water or wastewater using coagulants has been applied traditionally to reduce turbidity, by removing nonsettling or slowly settling (colloidal) solids from source waters.[15,16] The coagulation process promotes aggregation of the suspended solids to form flocs, which then can be removed through sedimentation and/or filtration. Dissolved inorganic contaminants, such as arsenic and other toxic metals can be also removed during coagulation treatment by alum or ferric chloride addition through the mechanisms of adsorption and occlusion.[17] During the adsorption process, the dissolved inorganic contaminant attaches to the surface of a particle, whereas occlusion occurs when the dissolved contaminant is adsorbed to a particle and then entrapped as the particle continues to agglomerate. Several studies have been reported for removing arsenic by coagulation, mainly from drinking water sources—most of them using $FeCl_3$[18–21] or alum,[19–21] whereas some researchers have examined the use of cationic polymers for enhanced removal of arsenic.[21] No citations exist in literature regarding the removal of arsenic by coagulation, enhanced by anionic polymers. Most of these studies have focused on achieving residual arsenic concentrations lower than the maximum concentration limit (MCL) of arsenic in drinking water ($10\,\mu g/L$), starting from initial concentrations rather low ($25–50\,\mu g/L$). This work has been focused on the treatment of industrial wastes containing arsenic ($100–1000\,\mu g/L$).

The main objectives of this work were to determine the efficiency of the examined modified treatment method (coagulation–pipe flocculation/direct filtration) for initial arsenic concentrations, which are typical of wastewaters, and to investigate the main parameters affecting arsenic removal, such as the type and the dosage of the coagulant (ferric chloride or alum), the type and dosage of the polyelectrolyte (cationic or anionic), initial arsenic concentration, and the effect of linear velocity, during the filtration step. Some of the experiments have been focused on treatment of lower arsenic concentrations (0.1–0.5) than the concentrations usually found in wastewaters (0.5–2 mg/L),[5] in order to evaluate the applicability of the modified technique for either wastewaters or drinking water. The experiments were performed in a laboratory scale.

The originality of this work was mainly based on the systematic investigation of coagulation/direct filtration, applying common coagulant agents enhanced by the presence of polyelectrolytes. It was an attempt to compare coagulation with ferric chloride or alum, enhanced with cationic or anionic polyelectrolytes in the same experimental unit leading to an integrated and comparable view of the effectiveness of coagulation/direct filtration as a treatment method for arsenic removal from industrial wastewaters. Furthermore, it has been an effort to modify the traditional coagulation/flocculation method, by using the "pipe flocculation" process. Flocculation in pipes represents a water treatment approach towards reducing overall flocculation process time as well as capital costs and space requirements.[22] The application of pipe flocculation, combined with the direct filtration step renders the applied technique quite interesting and promising, especially in cases where the need for removing contaminants on site is urgent.[22]

MATERIALS AND METHODS

Materials

Arsenate standards were prepared from $Na_2HAsO_4 \cdot 7H_2O$ (analytical grade) diluted with tap water of the following characteristics: conductivity $= 760\,\mu S/cm$, hardness $= 309\,\mu g/L$, and turbidity $= 0.3$ NTU; initial arsenic concentrations were in the range 0.1–1 mg/L. For the coagulation experiments, all solutions were prepared with tap water and all glassware were previously acid-washed. Stock solutions of $FeCl_3 \cdot 6H_2O$ (4000 mg/L) and $Al_2(SO_4)_3 \cdot 18H_2O$ (2000 mg/L) were prepared. The final concentrations of these reagents varied between 2 and 20 mg/L for iron (as ferric chloride) and between 4 and 10 mg/L for Al (as $Al_2SO_4 \cdot 18H_2O$). The polymers used were ZETAG-57 (cationic polyelectrolyte) and MAGNAFLOC-156 (anionic polyelectrolyte), obtained from Allied Colloids (Bradford, Yorkshire, UK). The examined concentrations of the polymers were in the range 0.5–5 mg/L for the anionic polyelectrolyte and 0.5–3 mg/L for the cationic one.

Methods

The applied treatment method, coagulation–direct filtration was based on a two-stage operation. Firstly, ferric chloride or alum was added to the arsenate-contaminated water and its coagulating action was enhanced by the presence of cationic or anionic polyelectrolytes. The next step involved the direct filtration of produced insolubles through a column, which contained a bi-layered sand bed. The pores of filter material were gradually clogging and the sand bed needed regeneration, performed by the backwashing action of a clean aqueous stream. The removed toxic sludge was collected and had to be disposed properly. The initial pH value of arsenic-contaminated water was the pH of the tap water—in this case 6.7. A schematic representation of this removal process can be seen in Fig. 1. The experimental set-up consisted of the

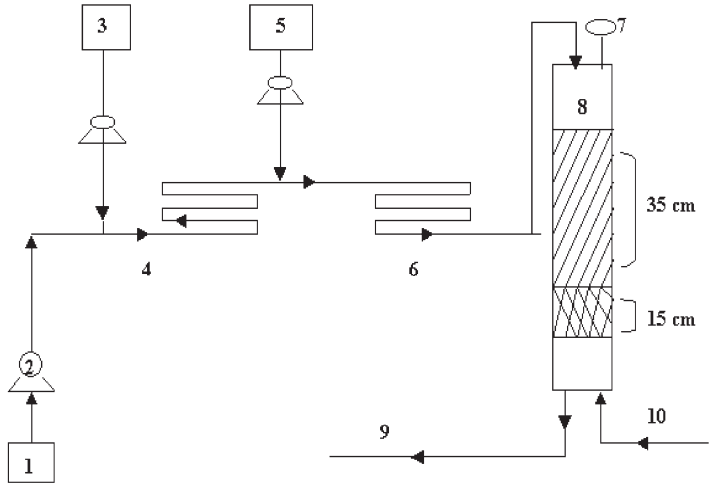

Figure 1. Schematic representation of arsenic removal process. 1: influent: As(V)-contaminated tap water, 2: peristaltic pump, 3: coagulant addition unit, 4: micro-flocculation pipe (0.6 m length, 15 mm inner diameter), 5: polyelectrolyte addition unit, 6: macro-flocculation pipe (0.6 m length, 7 mm inner diameter), 7: manometer, 8: bi-layered sand filter (particle size 0–2.5 and 4–8 mm), 9: effluent, 10: water for backwashing the sand filter. Column characteristics: column height: 1 m, sand bed height: 50 cm, inner diameter: 68 mm.

coagulant addition unit, the polymer addition unit, and the bi-layered sand filter. According to the process, the coagulant agent (ferric chloride or alum) was added to the arsenic-contaminated tap water. Following coagulant addition, “micro-flocculation” occurred within the connecting pipes and during the in-flow, as induced by the presence of a peristaltic pump. The next step involved the polyelectrolyte addition (anionic or cationic) and a second coagulating step “macro-flocculation” took place. It has been reported in literature citations that the flocculation inside the connecting pipes (pipe flocculation) is indeed significant and very efficient.[22] Then the flocculated aqueous stream passed through the filtration (Plexiglas) column, in a downflow operative mode. The insoluble products were filtered and the effluent was removed from the bottom of the bed. When the sand filter clogged, it was backwashed with tap water, while the toxic sludge was removed and the operation continued. The duration of each experimental run was almost 90 min. This time duration was found to be sufficient for satisfied arsenic removal to occur. Most of the experimental runs were performed at least three times and the average values were presented.

Arsenic Determination Methods

The determination of arsenic in the influent and effluent aqueous streams was performed by the molybdenum blue photometric method, for relatively high arsenic concentrations (>0.2 mg/L), in order to avoid subsequent dilutions and by the hydride generation–atomic absorption spectrometry (HG–AAS), for lower arsenic concentrations (<0.2 mg/L).

Molybdenum Blue Photometric Method

In this method, arsenic(V) reacts with ammonium molybdate to form a heteropoly molybdoarsenate, which is then reduced by a suitable reducing agent to form strongly colored "molybdenum blue," where the molybdenum is present in a lower valence. Then the molybdenum blue is measured photometrically at 660 nm and the calculation of arsenic concentrations is performed by means of the respective calibration curve. This method can be applied for the determination of arsenic concentrations between 0.2 and 4 mg/L, with experimental accuracy of $\pm 10\%$.[23]

Hydride Generation Coupled with Atomic Absorption Spectrometry

This analytical method was used for the determination of arsenic concentrations lower than 0.2 mg/L. The method is based on the reaction between inorganic arsenic and sodium borohydride to form volatile arsine. The created arsine is then detected by AAS and arsenic concentration is calculated by means of the respective calibration curve. The analysis was performed in a Perkin–Elmer 2380 AAS, coupled with a Perkin–Elmer MHS 10 HG unit (Perkin–Elmer Corporation, Norwalk, Connecticut). The detection limit of this method was found to be in the range between 0.001 and 0.02 mg/L, presenting an accuracy of $\pm 4\%$.[24]

RESULTS AND DISCUSSION

As(V) Removal During Ferric Chloride Coagulation

The effect of ferric cations addition on the removal of arsenic from contaminated aqueous streams was initially investigated. The results presented in Fig. 2 showed that there is a significant percentage of arsenic removal, during the treatment with ferric chloride. The simultaneous addition of polyelectrolytes, either cationic or anionic, along with ferric chloride was also examined. Blank

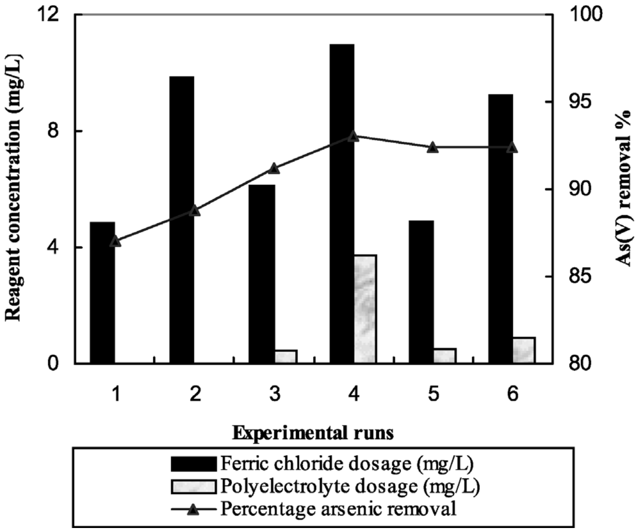

Figure 2. Arsenic removal by $FeCl_3$ coagulation enhanced with anionic or cationic polyelectrolytes; $[As]_o = 1\,mg/L$ (1 and 2 no polymer addition, 3 and 4 with cationic, and 5 and 6 with anionic polyelectrolyte addition).

experiments were also performed, without any addition of chemicals and arsenic removal was found to be less than 25%, probably due to some adsorption on the pyrite sand.

The addition of ferric chloride caused a substantial increase in the arsenic removal, but above a certain ferric chloride dosage, the increase in arsenic removal was not significant. Doubling of coagulant dosage resulted in a 2% increase in arsenic removal. On the other hand, improved arsenic removal was achieved by the addition of coagulant aids (polymers). The addition of cationic polymers enhanced the efficiency of the method, as the percentage arsenic removal was found to be much higher than the removal achieved without. The highest arsenic removal was found to be around 93%, when the concentrations of $FeCl_3$ and cationic polymer were 11 and 4 mg/L, respectively. The addition of anionic polymers led to similar results, increasing the arsenic removal up to 92%, under optimum $[FeCl_3]$/[anionic polymer] concentrations, which was found to be 9/0.9 mg/L. The addition of coagulants can facilitate the removal of arsenic from the aqueous stream, by converting the soluble As(V) species into insoluble products.

These products might form through precipitation, co-precipitation, or adsorption mechanisms.[20] As the formation of insoluble products was performed during the flow in the connecting pipes, the most probable mechanisms were adsorption and co-precipitation of inorganic arsenic oxyanions on iron oxides. The result was the formation of highly insoluble ferric arsenate ($FeAsO_4$); the solubility product of these species is quite low, being $10^{-20}\,mol^2/L^2$.[8] The application of polyelectrolytes resulted in the formation of greater particles,[21] which could be removed more efficiently through the sand filter. As far as it concerns the fact that cationic polyelectrolytes were proved more effective than anionic ones, this can be attributed to the fact that these polymers increase the cationic character of the inorganic coagulant, resulting in the formation of solids, which are more likely to adsorb oxyanions (arsenic).[22] The aforementioned results were considered as satisfactory, when compared with others mentioned in the literature,[19,21] as they were achieved from initial As(V) concentrations as high as 1 mg/L, which is typical of wastewaters contaminated with arsenic. Furthermore, in most of the previous studies the amount of coagulants applied were much higher[19–21] (20–30 mg/L) than the applied concentrations in this study (4–11 mg/L), indicating the significant improvement of the traditional coagulation treatment method, by applying the combination of pipe flocculation followed by direct filtration. The treatment technique showed greater efficiency, up to 99% As(V) removal, when lower initial As(V) concentrations were applied.

The effect of initial arsenic concentrations (0.1–1 mg/L) was subsequently investigated, by keeping constant the concentrations of coagulant and polymers at 9 and 0.9 mg/L, respectively, for both types of polyelectrolytes. These concentrations were chosen, because they corresponded to the optimum conditions for arsenic removal, when using the anionic polyelectrolyte. The objective of these experiments was to examine whether the treatment method was as efficient as when treating aqueous solutions of higher initial arsenic concentration (>1 mg/L), therefore, it could be applied to certain drinking water treatment problems. The results are shown in Fig. 3a and b, which represent the percentage arsenic removal as well as residual arsenic concentration.

In Fig. 3a, it can be noticed that the treatment of aqueous solutions, which contain arsenic at concentrations up to 0.5 mg/L, led to residual arsenic concentrations lower than 0.01 mg/L. These results refer to treatment by $FeCl_3$ and cationic polyelectrolytes. The addition of the anionic polyelectrolyte (Fig. 3b) was not proved to be equally efficient.

The effect of linear velocity was subsequently examined, as in processes where bed filtration is involved; linear velocity is a critical parameter affecting the overall applicability of the method. These experiments were performed using ferric chloride enhanced with anionic polyelectrolyte and the results are shown in Fig. 4, which indicate that linear velocity was found to affect the treatment efficiency. The best results were achieved at linear velocities around 5.5 m/hr; it was also shown that as linear velocity was further increased, the removal of As(V) was decreased, as at higher linear velocities, the sand bed was unable to

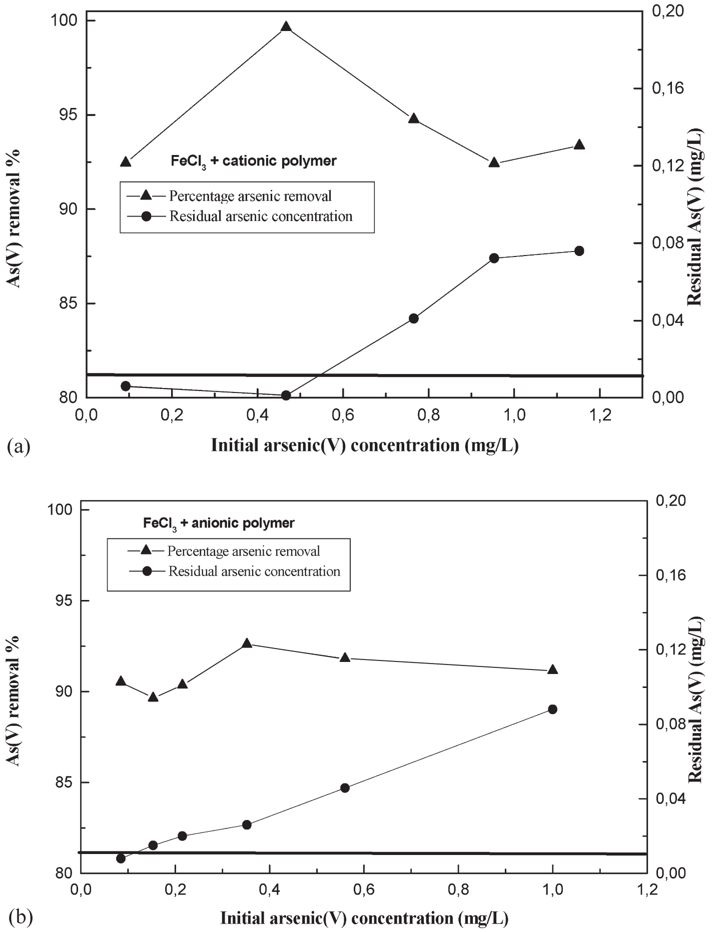

Figure 3. (a) Effect of initial arsenic concentration during $FeCl_3$ coagulation enhanced by the presence of cationic polyelectrolyte ($[FeCl_3] = 9\,mg/L$, $[poly^+] = 0.9\,mg/L$). (b) Effect of initial arsenic concentration during $FeCl_3$ coagulation enhanced by the presence of anionic polyelectrolyte ($[FeCl_3] = 9\,mg/L$, $[poly^-] = 0.9\,mg/L$).

filter all the produced particulates, or the time was not sufficient for flocculation to occur in the pipes. The examination of lower linear velocities (lesser than four) was not investigated, as it was considered that for lower linear velocities, the applicability of the method would be restricted.

Summarizing the results obtained by coagulation with ferric chloride, it can be observed that the addition of cationic polyelectrolytes enhanced the removal

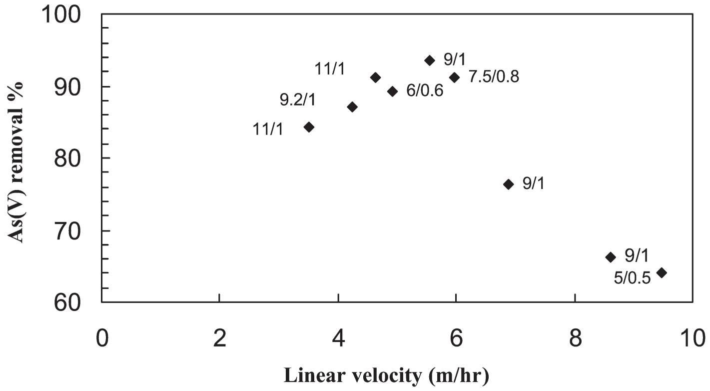

Figure 4. Effect of linear velocity on percentage arsenic removal for $[As]_o = 1\,mg/L$. These values represent the coagulant/polymer concentrations ($FeCl_3$/anionic), applied in each experimental run.

of arsenic. The treatment method, under optimum experimental conditions was proved to be effective for a wide range of initial arsenic concentrations, whereas the MCL for arsenic was achieved even for relatively high initial arsenic concentrations. These results are considered as more efficient regarding similar published papers.[18–21] On the other hand, the addition of anionic polyelectrolyte, although it enhanced arsenic removal to a certain extent was not found to be equally efficient, when treating aqueous solutions of lower arsenic concentrations.

The pH in all the experiments was kept at 6.7 (pH of tap water), in order to avoid further treatment of water. Moreover, literature citations[19] indicate that this is the optimum pH for As(V) removal during coagulation. At pH 6.7, iron cations are insoluble and the resulting solids are positively charged, whereas at higher pH values, above 10, iron is present as the monomeric form $Fe(OH)_4^-$,[25] which is ineffective for arsenic removal.

As(V) Removal During Alum Coagulation

In order to obtain an integrated view of the effectiveness of the modified treatment coagulation/direct filtration technique, the addition of aluminum sulfate (alum) as coagulant agent, enhanced with either cationic or anionic polyelectrolytes was subsequently examined. The results regarding arsenic removal, by varying the coagulant dosage as well as the type and dosage of the polyelectrolyte are presented in Fig. 5.

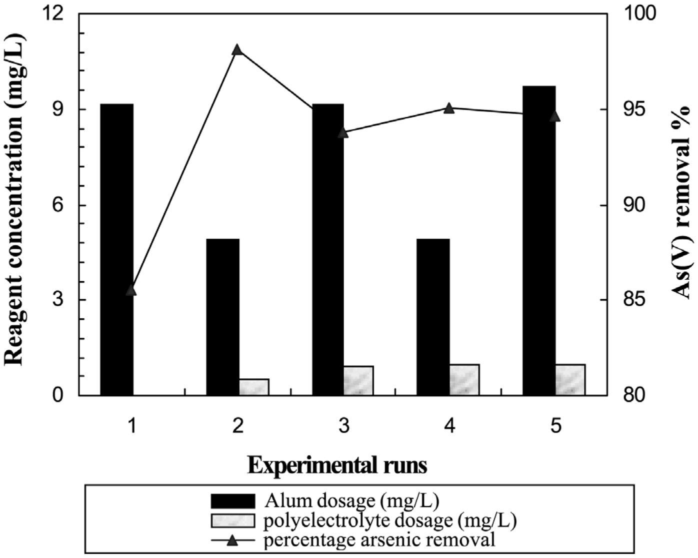

Figure 5. Arsenic removal by aluminum sulfate coagulation enhanced with cationic or anionic polyelectrolytes; $[As]_o = 1\,mg/L$ (1 no polymer addition, 2 and 3 with anionic, and 4 and 5 with cationic polyelectrolyte addition).

The results showed that coagulation with alum was found to be less efficient than iron coagulation, on a weight basis (i.e., mg/L $FeCl_3$ or alum). This difference could be attributed to incomplete precipitation of the added aluminum as the respective amorphous hydroxide solid.[19] When polyelectrolytes were added, either cationic or anionic, the removal results were increased. These results indicate that the use of cationic polyelectrolytes, as in coagulation with ferric chloride, proved to be more efficient than anionic because it increased arsenic removal up to 98%, whereas the addition of anionic polyelectrolyte led to maximum percentage removal of As(V) up to 94%. The presence of cationic polyelectrolyte in this case was found to present some limitations, as far as it concerns the maximum concentrations added; above a certain ratio of alum/cationic polymer, arsenic removal was found to decrease. This decrease in arsenic removal by the increase in alum dosage was also reported in similar published papers.[20]

The effect of initial arsenic concentration was also investigated for the case of alum coagulation. These results were obtained by varying the initial arsenic concentration from 0.1 to 1 mg/L, by adjusting the reagent concentrations [Alum]/[polyelectrolyte] = 9/0.9 mg/L, as in experiments performed by ferric chloride. This would enable the direct comparison between the results obtained by the two methods, which are shown in Fig. 6a and b.

From these figures it can be noticed that using either the cationic or anionic polyelectrolyte, the treatment method becomes more effective, when applied to aqueous solutions with low initial arsenic concentrations (<0.2 mg/L). The achieved residual arsenic concentration was lower than

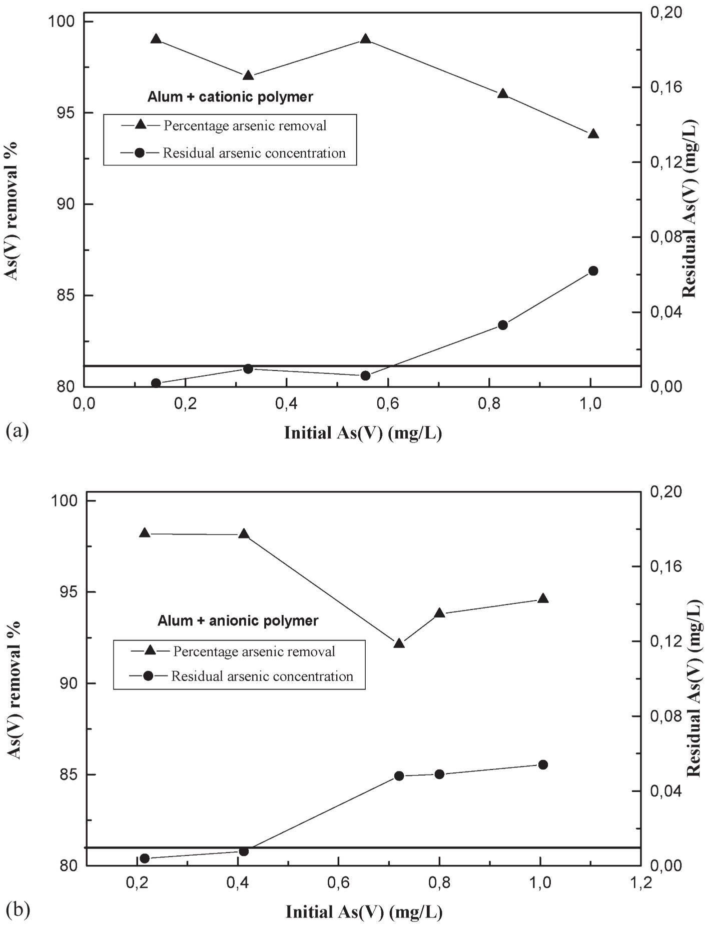

Figure 6. (a) Effect of initial arsenic concentration during alum coagulation enhanced by the presence of cationic polyelectrolyte ([Alum] = 9 mg/L, $[\text{poly}^{+}] = 0.9$ mg/L). (b) Effect of initial arsenic concentration during alum coagulation enhanced by the presence of anionic polyelectrolyte ([Alum] = 9 mg/L, $[\text{poly}^{-}] = 0.9$ mg/L).

the MCL for arsenic in drinking water (0.01 mg/L) and the upper concentration for achieving that limit was found to be 0.6 mg/L of As(V) for the case of cationic polyelectrolyte and 0.4 mg/L for the case of anionic one. The cationic polyelectrolyte was again proved to be slightly more effective in removing arsenic than the anionic one and this can be attributed to the same reasons as in coagulation with ferric chloride.

The above results were considered as very satisfactory, as in most published papers about arsenic removal with alum coagulation, the arsenic removal efficiencies were less than 90% and for initial arsenic concentrations much lower than 1 mg/L. The application of pipe flocculation in combination with direct filtration proved to be very efficient, as in some cases arsenic was removed by 99%.

CONCLUSIONS

In summarizing the obtained experimental results, certain conclusions can be drawn. The modification of the conventional coagulation–flocculation technology, by applying the pipe flocculation process was found to be very efficient for the removal of arsenic anions, from wastewaters and can find applications in drinking water treatment also. The method was efficient for both iron and alum coagulation and both types of polymers (cationic or anionic) were found to increase the overall efficiency of the treatment method. This conclusion is interesting, as until date, no researcher has used anionic polyelectrolytes for the removal of arsenic. The treatment technique presents several advantages towards conventional coagulation processes, as during pipe flocculation the overall flocculation process time was decreased as well as capital costs and space requirements. It is a treatment option, which can contribute to the overall body of knowledge, as it shows that coagulation/direct filtration can be used for the removal of high arsenic quantities from both wastewaters and drinking water. It was found that in almost all cases, the arsenic concentration can be reduced down to 10 μg/L from initial concentrations of over 400 μg/L. Therefore, the application of this modified treatment method does not require any additional treatment of waters or wastewaters, regarding the removal of arsenic.

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