Pre-disinfection columns to improve the performance of the direct electro-disinfection of highly faecal-polluted surface water

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Abstract

In this work, it is faced the design and evaluation of a new concept of pre-disinfection treatment, especially suited for highly-polluted surface water and based on the combination of coagulation-flocculation, lamellar sedimentation and filtration into a single column unit, in which the interconnection between treatments are important parts of the overall process. The new system, so-called PREDICO system (PRE-DI disinfection Column), has been built with low-cost consumables from hardware store (in order to promote its in-house construction in poor countries) and it has been tested with a mixture of 20% raw wastewater and 80% surface water (in order to simulate an extremely bad situation). Results have confirmed that it helps to avoid fouling in later electro-disinfection processes, while it also attains a very remarkable disinfection (3-4 log-units), which can be added to the removal of pathogens attained by the electrolytic cell (more than 4-log units). The most important sizing parameters for the PREDICO are the surface loading rate (SLR) and the hydraulic residence time (HRT) and values of SLR under 20 cm min⁻¹ and of HRT over 13.6 minutes in the PREDICO system are suitable to warrant an efficient performance of the system.
Keywords

Process integration; electro-disinfection; diamond electrodes; pre-treatment; coagulation; lamellar decanter

Highlights

− Pre-treatments help to prevent fouling in later electro-disinfection processes
− Very remarkable disinfection of highly-polluted water (3-4 log-units)
− Pre-treatments can be combined into a single column unit with an in-house design
− The pre-disinfection column can be easily integrated with electro-disinfection

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1. Introduction

Disinfection of highly polluted surface water is an important challenge for Society (Ngwenya et al., 2013; Dewil et al., 2017). In fact, it is one of its main achievements of the last century and it has helped to increase the average life expectancy in more than double in developed countries (Bebelis et al., 2013). Unfortunately, it does not arrive to all regions of the World in the same extent and there are many places in which it application is still insufficient. In addition, the scarcity of water in many places of the World (intensified by the important effects of the climate change) makes very difficult to find good reservoirs very often. The worst situation is when both problems are combined and a good example of this combination is found in rural communities in southern Africa, where often the only possible drinking supply does not contain high quality water and lack of cheap technologies generates a very serious social and health problem.

Disinfection is a key step to provide safe drinking water (Bergmann and Koparal, 2005b). This treatment consists of killing pathogens contained in water and it is less exigent than sterilization which means total depletion of microorganisms from water.

There are many technologies applied currently to disinfect water (Okafor, 2011). Most of them show important drawbacks, not only related to economy but also to the formation of hazardous species. For this reason, research on the topic is very active nowadays and there are many processes in development. Among them, electro-disinfection is gaining more and more attention every day (Martinez-Huitle and Brillas, 2008; Bebelis et al., 2013; Radjenovic and Sedlak, 2015; Sarkka et al., 2015).

Nowadays, there are many electrochemically-based processes in use (mainly with Mixed Metal Oxides anodes) (Bergmann and Koparal, 2005a; Henquin et al., 2013; Mezule et al., 2015; Cotillas et al., 2016a; Valero et al., 2017) and in evaluation (mainly with
diamond anodes) (Bergmann et al., 2002; Cano et al., 2012; Rajab et al., 2015; Schaefer et al., 2015; Cano et al., 2016; Cotillas et al., 2016b). One of them is the CabECO ® process, which is based on the use of an electrochemical cell capable to produce high amounts of ozone (among other disinfectants), even in low conductivity water (Fryda et al., 2016). This technology has also been applied successfully to the direct disinfection of water, showing a very interesting performance even in the treatment of strongly polluted water and attaining easily killing rates of more than 3-log units (Isidro et al., 2018).

Presence of solids in water may become a handicap for the direct electrochemical disinfection because in order to obtain highly efficient processes the inter-electrode distance should be kept as low as possible in order to obtain low cell voltages and high current intensities (Rodrigo et al., 2010). These solids can obstruct the flow of water or even they can contribute to the fouling of the electrode surface preventing electrochemical reactions (Buschini et al., 2004). For this reason, it is necessary to combine the electrochemical disinfection with pre-disinfection treatments capable to give reach high quality water to be disinfected (Zaleschi et al., 2012; Zaleschi et al., 2013).

Hence, the basic idea aimed in this paper is to combine into a single process all the unit operations required to pre-treat highly-polluted surface water before disinfection, in order to be able to obtain water with quality high enough to be connected to a CabECO ® cell protecting the surface of the electrodes and avoiding the blocking of the electrochemical cell. A comparison of disinfection results, with and without this pre-treatment, and some preliminary sizing details of the new technology are given in this work.

2. Material and Methods

PREDICO system. Typically, in most municipal water treatment facilities, the treatment of surface water consists of a sequence of coagulation-flocculation, sedimentation,
filtration and disinfection (Boyjoo et al., 2013; Nakazawa et al., 2018; Sillanpaa et al., 2018). The in-house made PRE-DIIsinfection Column here introduced (so-called PREDICO system), combines the three first units into one, by sequencing them into a column in which the top of a unit becomes the bottom of another one, trying to get advantages of the gravity driving force. Then, the bottom of the pre-disinfection column consists on a coagulation chamber that simultaneously acts as the concentration zone of the lamellar decanter, while the top of the lamellar decanter acts as a reservoir for the granulated activated carbon (GAC) filtering. Thus, the PREDICO process tries to combine all the treatments included in a typical Surface Water Treatment Facility into an in-house made single column and makes use of the connection to increase the performance of each unit (Figure 1). This functionality of the connections between units is a very efficient approach, previously used successfully in other water treatment technologies (Ayekoe et al., 2017).

Regarding the setups used in this work, its core design has been carried out looking for an easy replication. The raw water is introduced at the bottom in the coagulation-flocculation chamber (E01) which is initially sized for a hydraulic retention time of 6 minutes and where mixing is provided by the inlet flow of water which is entered in the bottom of the chamber (see details in Figure 2). After that the water goes through a lamellar decanter (E02) that is coupled with the first equipment and has a total HRT of 10.3 minutes and a superficial load of 20 cm min⁻¹. Finally, in the vertical direction there is a filtration equipment (E03) with a bed of Granular Activated Carbon (GAC) sized for a hydraulic residence time of 2.0 minutes. This system is completed with a pump B01 and a flow meter (F01) to measure the treated water flow rate. Two units were constructed, one for 20 L/h (bench scale plant) used to obtain the preliminary sizing data required for a larger prototype sized for 300 L/h (pilot plant). In the pilot plant, the lamellar decanter consists of 71 thinner pipes (DN20) inside the big pipe (DN190) that conform the column.
A picture showing both is shown in Figure 1. One advantage looked for in its design is that it can be easily made in-house and, hence, it can be easily and economically applied in rural communities of poor countries by using common hardware store materials (mainly pipes of different diameters).

In the evaluation of the PREDICO system, the key parameter is the surface loading rate (SLR) which is a velocity (cm min\(^{-1}\)) also referred to as the surface settling rate or surface overflow rate. This parameter can be calculated as the ratio between the flowrate and the flow-through section of the decanter or the filter. This parameter can be used for the scale-up of the separation units and in this work, tests are carried out to confirm this point by varying the flowrate that pass through the PREDICO.

In addition, and taking into account that PREDICO does not only include separation units (sedimentation and filtration) but also reactive units (coagulation-flocculation), it is also important the hydraulic residence time (HRT), which can be calculated as the ratio between the flowrate and the volume of the reaction unit.

It is important to highlight that in our system, in order to use a single value (because there are very different free surface areas), the arbitrary value of the total outer pipe was used for the calculations.

**Electrolytic system.** Electrolysis tests were carried out in a CabECO® cell delivered by CONDIAS (Itzehoe, Germany). It is a cell specially designed to produce ozone in low-conductivity water. It contains four DIACHEM® electrodes, made on a substrate which consists of a niobium mesh (type B). The electrodes (50x24x1.3 mm\(^3\)) are assembled in two stacks with a NAFION® cation exchange membrane separating the anode and cathode and acting as the electrolyte. The cell was connected to the water reservoir by a peristaltic pump and powered by a Promax DC FA-376 power supply. A Keithley 2000 multimeter was used to monitor current and cell voltage.
Fig. 1. Layout of the process (left). Picture (right) with the bench scale (front side) and pilot plant (rear side).

Fig. 2. Coagulation chamber of the pilot plant under no flow where sludge is settled (left) and flow conditions (right) where the sludge is suspended.

3. Results and discussion

Direct disinfection using electrochemical technology is expected to undergo severe problems during the treatment of highly-polluted surface water, with biofouling being the
most important. This problem leads to a very important decrease in the efficiency of the disinfection process. This is confirmed in Figure 3, which shows the effect of the current intensity (I) on the concentration of total aerobic microorganisms and total coliforms during the treatment with a CabECO® unit of a real highly polluted surface water (made by merging 20% wastewater with 80% surface water). At this point, it is important to take into account that total aerobic microorganisms is a parameter that quantifies pathogens and non-pathogens microorganisms and, hence, it gives information about the potential use of the technology for sterilization. On the contrary, total coliforms is the most applied indicator of fecal pathogens and it gives valuable information about the use of the technology for disinfection, in particular in the case of fecal polluted water. In both cases, the graph shows the ratio between the concentration of each type of microorganisms at the outlet (N) and inlet (N₀) of the electrochemical cell and results are plotted in log scale.

As it can be observed, sterilization and disinfection reached depends on the water flowrate treated and current intensity applied and results are not as good as those which can be obtained during the sterilization and disinfection of water with the CabECO® technology when the technology is applied to water with lower content of solids, for which this technology is able to attain more than 3-logs of disinfection even for very high flowrates (Isidro et al., 2018). Thus, the direct treatment of this highly-polluted water sample with the CabECO® leads to bad results in terms of killing microorganisms with maximum rates of only 1 log-unit, much below the values that typically are reached with other diamond electrolysis shown in the literature (Cano et al., 2011; Cano et al., 2012; Lacasa et al., 2013; Cano et al., 2016). In fact, this fouling also helps to explains that performance at higher flowrate was much better than at lower flowrates (despite the higher volumes of water treated) because of the shear stress caused by the flowrate on the surface of the electrodes, which contributes to the cleaning of the surface of the electrodes. This effect was also observed in other works in which surface-based treatment technologies were
applied to highly-polluted water (Zouboulis et al., 2014; Julio et al., 2015). Anyway, one log-unit can be attained in the removal of total aerobic microorganisms and slightly over 1 log-unit in the case of total coliforms when the high flowrate is treated. At this point, in comparing the two microbiological indicators it can be stated that both, sterilization and disinfection, perform with very similar efficiency and no remarkable differences can be pointed out.

**Fig 3.** Influence of the current intensity applied on the removal of microorganisms by direct application of the CabECO® technology to fecal-polluted water (20% wastewater with 80% surface water), expressed as the ratio between the microorganisms contained in the effluent of the cell (N) and the microorganisms contained in the inlet (N₀). ▲ Total aerobic microorganisms, treated flowrate 150 L/h; △ Total aerobic microorganisms, treated flowrate 300 L/h; ● total coliforms, treated flowrate 150 L/h; ○ total coliforms, treated flowrate 300 L/h.

These results clearly pointed out that it is necessary the attachment of a pre-disinfection treatment unit in order to improve the results of the electro-disinfection. This necessity of pre-treatment operations is not only found for the electrochemical technologies and it has
been used in many other water treatment approaches (Pio et al., 2015; Im et al., 2018)). Initially, this complementary treatment can be an advantage for the CabECO® because in addition to avoid the formation of biofilm on the surface of the electrodes, it can help to reduce significantly the concentration of pathogens to be removed and it can help to attain the disinfection of the water.

In order to characterize the performance of the compact coagulation-decanter-filtration pre-disinfection column unit (PREDICO), a first point to be clarified is the effect of the flowrate, which is particularly important in the sedimentation stage of the unit. Thus, the flowrate should result in an ascending flow rate of flocs below the surface loading rate (SLR) in order to avoid loses of solids formed during the coagulation from the lamellar sedimentation stage of the PREDICO unit. Taking into account that solids to be settled correspond to the effluent of a coagulation with iron reagent (hence, iron hydroxide is the primary species in their composition), a first study was carried out in a bench scale plant in order to determine the maximum admissible SLR that should be applied to the lamellar settler to retain efficiently the solids produced during the coagulation stage. Results are shown in Figure 4.

![Figure 4](image_url)

**Fig 4.** Influence of the surface loading rate on the performance of the lamellar decanter contained in the PREDICO system in tests carried out at bench-scale. a) Total suspended solids in the clarified liquid and in the inlet of the GAC filter unit (empty for these experiments) in the bench scale plant. △ Position 1 (M-05 in Figure 1); ○ Position 2 (M-
04 in Figure 1); b) Total suspended solids at the bottom of the decanter (coagulation reactor): □ Position 3(M-03 in Figure 1); ◊ Position 4(M-02 in Figure 1).

As it can be seen, as the water flow increases, quality of the effluent becomes worse. In fact, operating under 20 cm min⁻¹ seems to be a good practice in order to prevent solids escaping from the unit. Regarding concentration of coagulant sludge at the bottom of the lamellar decanter (which in fact is the coagulation-flocculation chamber), it can be seen that as the flowrate increases, the sludge blanket becomes larger and solids become less concentrated. This may have a negative impact on the flocculation results because of the lower possibilities of collision between flocs. However, by working at fluid velocities under 20 cm min⁻¹ the concentration is over 1 g dm⁻³, hence high enough to assure good flocculation results (Zaleschi et al., 2012).

Once clarified the range of flow velocity through the system, a prototype (pilot plant) was sized for 300 L/h and constructed and additional tests were carried out at the pilot plant with flowrates ranging from 50 to 300 L/h in order to check if the scale-up was properly carried out. This pilot plant was sized so that the fluid velocity thought the cell (SLR) at the maximum flowrate was below 20 cm min⁻¹, hence avoiding loses of solids from the decanter and helping to enlarge the service lifetime of filters. Figure 5 shows the iron measured at different points of the setup during initial coagulation test. Results obtained confirm that particles are kept in the system and that only negligible concentrations of iron are lost during the treatment. Therefore, the use of the SLR as scale-up parameter for the separation units of PREDICO is confirmed with a scale-up ratio of 15 (20 L/h in the bench scale plant vs 300 L/h in the pilot plant). Likewise, it is confirmed how sludge is concentrated in the coagulation chamber of the pilot plant (which simultaneously acts as the bottom of the lamellar decanter) allowing a good performance of the flocculation
because of the mixing conditions attained and the promotion in the concentration of the flocs.

**Fig 5.** Influence of the surface loading rate on the performance of the lamellar decanter contained in the PREDICO system in tests carried out at pilot plant scale. a) Iron measured by ICP in the clarified liquid and in the inlet of the filter unit (empty for these experiments) in the pilot plant △ Position 1 (M-05 in Figure 1); ○ Position 2 (M-04 in Figure 1); b) iron measured at the bottom of the decanter (coagulation reactor) □ Position 3 (M-03 in Figure 1); ◊ Position 4 (M-02 in Figure 1)

The coagulation of colloids and the removal of solids by sedimentation and filtration is expected to help to remove microorganisms from water as it was pointed out in previous works of our group (Zaleschi et al., 2012; Zaleschi et al., 2013) and of other researchers (Chon et al., 2014; Kpan et al., 2017). Thus, the highly fecal-polluted water treated made by merging 20% raw wastewater with 80% surface water was passed through the PREDICO system at different flowrates and the removal of microorganisms at the inlet and outlet was measured. Figure 6 shows changes observed in the steady state value reached in each test in which flowrates from 50 to 300 L/h were passed through (resulting in HRT in the coagulation chamber ranging from 4.5 to 27.2 minutes).
Fig 6. Influence of the hydraulic retention time on the removal of microorganisms by direct application of the PREDICO technology to fecal-polluted water (20% wastewater with 80% surface water), expressed as the ratio between the microorganisms contained in the effluent (N) and influent (N₀) of the PREDICO system (pilot plant). △ Total aerobic microorganisms; ○ Total coliforms

As it can be seen, there is a depletion of microorganism (sterilization) of more than 3-log units and of pathogens of more than 4-log units (disinfection) and efficiency depends on the hydraulic retention time in the coagulation chamber. Up to 13.6 minutes, there is a clear log-decrease of the microorganisms’ concentration. From that HRT, the maximum disinfection capacity is reached and no improvements were found. At this point, the concentration of microorganisms in the effluent of the treatment is really low, but it has to be taken into account that this PREDICO process does not kill microorganisms but simply concentrated them into the iron hydroxide sludge, so it is necessary to purge this sludge and to kill microorganisms in a later stage (Zaleschi et al., 2012; Zaleschi et al., 2013). This can be easily done simply by acidifying the sludge in order to solubilize iron and use this iron again in the system to avoid the use of new iron once neutralized.
As the concentration of microorganisms at the outlet of the PREDICO unit was very low, preliminary test for the evaluation of the CabECO® technology was not completely successful in terms of characterization of the technology (total depletion of microorganism was obtained but initial concentrations were rather low, so no relevant conclusions were found). Hence, in order to check the capability of the CabECO® process to disinfect highly microbial loaded water, the outlet of the PREDICO system was intensified with coliforms, pseudomonas and total aerobic microorganism by adding Pseudomonas Aeruginosa and E. Coli from pure cultures, up to concentrations similar to those contained in the non-pretreated water. Figure 7 shows the effect of the current intensity and flowrate on the steady state disinfection attained by the CabECO® in this case.

**Fig 7.** Influence of the applied current intensity on the removal of microorganisms by direct application of the CabECO ® technology to water previously treated by the PREDICO system and intensified with microorganisms to reestablish the initial values contained before the PREDICO. N and N₀ represents the microorganisms contained in the effluent and influent of the electrolytic system. a) disinfection: ● total coliforms, treated flowrate 150 L/h; ○ total coliforms, treated flowrate 300 L/h; ■ P. aeruginosa, treated flowrate 150 L/h □ P. aeruginosa, treated flowrate 300 L/h. b) sterilization: ▲ Total aerobic microorganisms, treated flowrate 150 L/h; △ Total aerobic microorganisms, treated flowrate 300 L/h.
As observed, results are much better than those obtained during the treatment of the raw water (before application of the PREDICO process) and in this case the effect of the flowrate is just as expected and, hence, higher disinfection rates are attained when treating lower flowrates of water. Obviously, no fouling was observed on the electrode surface after the treatment and hence, in addition to the 3-4 log units of disinfection that the PREDICO system can attain, up to 3-log units in total aerobic microorganisms (sterilization) and 4-log units in coliform (disinfection) can be attained, assuring the complete treatment of almost any type of water with the combination of the PREDICO and the CabECO® technologies. Then, from the results obtained in this work, it can be stated that all together, the combination of the pre-disinfection and the disinfection units, is a promising and feasible choice because it guaranty a highly efficient treatment, capable to improve very efficiently the quality of very poor water.

4. Conclusions

An efficient system, so-called PREDICO system (PRE-Disinfection Column), has been developed and evaluated in this work. In operating this system with a SLR under 20 cm min⁻¹ and of HRT over 13.6 minutes, PREDICO can reach more than 3-log units removal in total aerobic microorganisms (sterilization) and 4-log units in coliform (disinfection) and prevent fouling of the later electro-disinfection cell when treating a strongly polluted surface water (made of by mixing 80% surface water with 20% raw urban wastewater). Results are additive to those obtained by the electrolytic disinfection (more than 4-log units) and results in a highly efficient combined treatment. PREDICO has been carefully designed so that it can be easily and economically applied in rural communities of poor
countries by manufacturing and replicating it using common hardware store materials (mainly pipes of different diameters).

Acknowledgments

This research belongs to the SafeWaterAfrica project, funded by the European Union's Horizon 2020 research and innovation programme under grant agreement No 689925.

References


