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Resumen	El presente artículo muestra los resultados de un estudio que se llevó a cabo para evaluar el efecto de la calidad del agua en la remoción de sílice disuelto mediante un proceso de electrocoagulación utilizando electrodos de aluminio. El sílice se encuentra en el agua de repuesto (RW) y de purga de las torres de enfriamiento (CTBW). Las pruebas se hicieron a escala semipiloto a flujo continuo en un tren de tratamiento consistente de electrocoagulación (EC), floculación, sedimentación y filtración en arena.
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Electrocoagulation to Remove Silica from Cooling Towers Water

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Abstract

VILLEGAS-MENDOZA, I. E., MARTÍN-DOMÍNGUEZ, A., PÉREZ-CASTREJÓN, S. & GELOVER-SANTIAGO, S.L. Electrocoagulation to Remove Silica from Cooling Towers Water. *Water Technology and Sciences* (in Spanish). Vol. V, No. 3, May-June, 2014, pp. 41-50.

This paper presents the results of a study carried out about the effect of water quality on the removal of dissolved silica using an electrocoagulation process with aluminum electrodes. Silica is found in replacement water (RW), usually known as make up water, and in cooling tower blowdown water (CTBW). Tests were conducted on a small pilot scale (~2 lmin^{-1}) with a continuous flow device. The treatment train consisted of electrocoagulation (EC), flocculation, sedimentation and sand filtration. Two distinct RW and two CTBW with different physicochemical characteristics were studied. The response variables analyzed were: efficiency of aluminum to remove silica (ratio mgl^{-1} of dosed Al^{3+}/mgl^{-1} SiO_2 removed), removal efficiency of dosed Al^{3+}, hydraulic head loss throughout the electrochemical reactor and voltage. The cost of the treatment for the four types of water is discussed. The ratio mgl^{-1} Al^{3+} dosed /mgl^{-1} silica removed ranged from 1.09 ± 0.06 to 1.33 ± 0.05 when treating RW and 0.85 ± 0.1 when treating CTBW. The consumption costs of energy, chemicals and electrodes for RW treatment ranged from US$ 0.52 to 0.74 m^{-3}, and was approximately US$0.53 m^{-3} for CTBW.

Keywords: Aluminum, blowdown, cooling tower, electrocoagulation, make-up, silica, water quality, concentration cycles.

Resumen

VILLEGAS-MENDOZA, I. E., MARTÍN-DOMÍNGUEZ, A., PÉREZ-CASTREJÓN, S. & GELOVER-SANTIAGO, S.L. Electrocoagulación para remover sílice en agua de torres de enfriamiento. Tecnología y Ciencias del Agua. Vol. V, núm. 3, mayo-junio de 2014, pp. 41-50.

El presente artículo muestra los resultados de un estudio que se llevó a cabo para evaluar el efecto de la calidad del agua en la remoción de sílice disuelto mediante un proceso de electrocoagulación utilizando electrodos de aluminio. El sílice se encuentra en el agua de repuesto (RW) y de purga de las torres de enfriamiento (CTBW). Las pruebas se hicieron a escala semipiloto a flujo continuo en un tren de tratamiento consistente de electrocoagulación (EC), floculación, sedimentación y filtración en arena. Se estudiaron dos RW y CTBW, con características fisicoquímicas diferentes. Las variables de respuesta analizadas fueron las siguientes: eficiencia del aluminio para remover sílice (relación mgl^{-1} de Al^{3+} dosificado/mgl^{-1} de sílice removido), eficiencia de remoción de Al^{3+} dosificado, pérdida de carga hidráulica a través del reactor electroquímico y el voltaje. Se calculó el costo del tratamiento de los cuatro tipos de agua. La relación mgl^{-1} de Al^{3+} dosificado/mgl^{-1} de sílice removido osciló de 1.09 ± 0.06 a 1.33 ± 0.05 al tratar RW, mientras que para CTBW fue de 0.85 ± 0.1. Los costos de energía, sustancias químicas y consumo de electrodos para el tratamiento de RW osciló de US$ 0.52 a US$ 0.74 m^{-3}, y el costo del tratamiento de CTBW fue de aproximadamente US$ 0.53 m^{-3}.

***Palabras clave**: aluminio, purga, torre de enfriamiento, electrocoagulación, agua de repuesto, sílice, calidad del agua, ciclos de concentración.*

Introduction

One of the most effective strategies to reduce water demand in industries is to reduce replacement water or make up water (RW) consumption in cooling towers (CT), since these equipments consume large amounts of water (Hinrichs and Kleinbach, 2012). The high concentration of silica (up to 100 mgl^{-1}) contained in groundwater commonly used as RW in certain regions throughout Mexico and Latin America (Demadis and Neofotistou, 2004), causes that the CTs operate at low concentration cycles. Cycles of concentration (CC) is the number of times that a chemical species of reference (in this case the silica), can increase its concentration before

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a portion of the water is discarded to avoid precipitation in heat transfer equipment. If X is a reference substance, then:

$$CC = (\text{concentration of X in blowdown} / \text{concentration of X in makeup water}) \quad (1)$$

RW in a CT can be calculated using Eq. (2), if the drift (figure 1) is considered near to zero

$$RW = E*CC/(CC-1) \quad (2)$$

Where E is the amount of evaporated water and CC represents the concentration cycles

On the other hand, cooling tower blowdown water (CTBW) can be estimated with Eq. (3)

$$CTBW = RW/CC \quad (3)$$

According to these equations, if CC increases, the required RW diminish and so does CTBW (Seneviratne, 2007). Figure 1 illustrates the main currents in a CT.

Several treatment methods exist to remove silica; however, the most common one has been chemical coagulation (ChC). This treatment is efficient in the removal of soluble and colloidal silica (Sheikholeslami and Bright, 2002), but has inherent problems in the cost, maintenance and production of sludge (Emamjomeh and

Figure 1. Mass balance of water for a cooling tower.

Sivakumar, 2009). Chuang *et al.* (2006) reported that the removal efficiency was about 7.4 mg Al_2O_3/mg SiO_2 when dosing poly-aluminium chloride (PACl) or alum in a range from 30 to 150 mg/L as Al_2O_3, this represents a 4:1 (aluminium:silica) mass relationship. There are other methods that can remove silica efficiently; these include nanofiltration, reverse osmosis, ion exchange and electro-deionization. Nevertheless, the costs of these methods are higher than those for chemical methods (Zeng *et al.*, 2007).

Electrocoagulation (EC) is an electrochemical technique that has been suggested as an alternative to conventional coagulation, and is characterized by its low sludge production, no addition of chemicals and easy operation (Emamjomeh and Sivakumar, 2009; Pérez-Castrejón *et al.*, 2012). The main difference between EC and ChC is the manner in which metal ions are added to water. EC produces flocks of metal hydroxides by electro-dissolution of soluble anodes, usually made up of iron or aluminium, while ChC promotes the formation of hydroxides by using metal salts such as aluminium sulfate or ferric chloride (Comninellis and Chen, 2010).

Several studies using EC to remove silica have been carried out: Den and Wang (2006); Kin *et al.* (2006); Wang *et al.* (2009); Liao *et al.* (2009); Schulz *et al.* (2009); however, only the last two authors worked with representative cooling tower water. On the other hand, up to our knowledge, there are not studies where the convenience of treating replacement water with respect to treat cooling tower blowdown water is analyzed.

This paper presents the results of treating make-up water from wells (RW) and cooling tower blowdown water (CTBW) at a pilot small scale. The results were used to determine the technical and economic feasibility of treating RW or CTBW in an EC process combined with a conventional clarification system for silica removal.
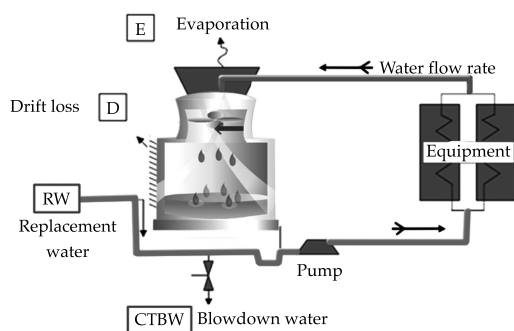
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Methodology

Type of water used

The water used in this study was obtained from two water supply wells (RW1, RW2); the water quality is different as they come from two distinct locations. On the other hand, two different blowdown cooling tower samples coming from two different industries (CTBW1, CTBW2) were the other studied matrices. The relevant quality parameters of the four sources of water are shown in table 1.

The RW1 matrix has been extensively studied by our working group under different conditions of operation, consistent previously obtained results ensures us that the system is stable and that the results are statistically reproducible (Villegas-Mendoza, 2009, 2011; Gelover-Santiago *et al.*, 2012). RW2 and CTBW1 come from the same cooling tower.

Description of experimental equipment and response variables

The experiments were carried out using a pilot small scale EC system, with a plug flow electro-chemical reactor attached directly to a three-stage mechanical flocculator followed by a high rate settler and a gravity-fed sand filter.

The reactor was made from acrylic having the shape of a rectangular parallelepiped with a dimension of height: 10 cm, length: 78 cm and width: 15 cm. Placed inside the reactor were 60 aluminium electrodes measuring 8 x 4 x 0.5 cm each, connected like deflecting screens in a monopolar arrangement (figure 2), forcing the water to behave like a flow plug throughout the reactor. The space between the electrodes was 0.6 cm and the total active anodic area of the electrodes was 0.19 m². The hydraulic head loss in the reactor was measured by a mercury differential manometer, allowing an evaluation

Table 1. Quality of analyzed water.

	RW1	RW2	CTBW1	CTBW2
pH	7	8.64	8.6	7.13
Conductivity (µS/cm)	200	795	1 625	1 290
Silica (mgL⁻¹)	50	83	164	195
Sulfates (mgL⁻¹)	25.2	83	250	298
Chlorides (mgL⁻¹)	8.38	43.1	111	338
Turbidity (NTU)	0.05	0.05	14	35.1

Figure 2. Electrochemical reactor.

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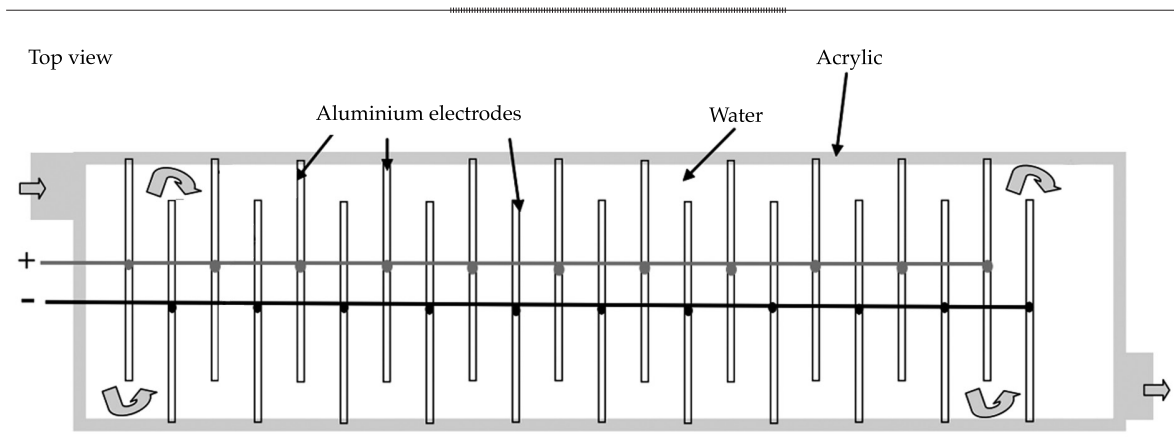Tecnología y

of the blockage caused by the formation of deposits on the surface of the electrodes.

The electrical power required to establish the desired theoretical concentration of Al^{3+} in each one of the experiments was calculated using Faraday's law (Comninellis and Chen, 2010; Pérez-Castrejón *et al*. 2012). It was supplied using a Sorensen DLM 40-15 model power source and the polarity of the electrodes was shifted every hour. The current density and flow rate of operation were 71 Am^{-2} and 1.7 $Lmin^{-1}$ respectively. In order to remove deposits formed during the preceding tests, at the beginning of each experiment the reactor was chemically washed with a solution at 30 % of commercial product with hydrochloric acid as the active ingredient. The response variables were:

- Efficiency of aluminium to remove silica (mgl^{-1} dosed Al^{3+} / mgl^{-1} of removed silica), after settler and filter.
- Efficient removal of dosed aluminium, after settler and filter.
- Hydraulic head loss (indirect measurement of the obstruction by deposits in the reactor).
- Voltage variation (indirect measurement of the passivation of the electrodes).

The data of silica removal efficiency were statistically analyzed. First of all, the data for RW1 were compared with data previously published by our group (Gelover-Santiago *et al.*, 2012), for the same water matrix (RW1) using the same pilot plant, aluminium cathodes and anodes, applying similar current densities and switching the polarity during the test. After this, a Student´s t-test for media comparison was applied to contrast the values for silica removal for the four types of water considered.

Analytical determinations

The concentration of silica and aluminium was determined by colorimetry with a DR/2010 Hach spectrophotometer by the Aluver and Molybdate methods, respectively. The pH was determined with an Orion 420A potentiometer model and the conductivity with an Orion 145 Model.

The film in the passivated electrodes was characterized using a X-Ray diffractometer Rigaku DMAX-2200 with a radiation K-alfa of cupper. Intensities were measured in the 2θ range between 3º to 100º, with a two theta step of 0.02 º and a scanning velocity of 1º/min 36 kV and 30 mA. The identification of crystal phases was carried out with the software Jade 6.5 and the database of the powder diffraction patterns (PDF) of the International Centre for Diffraction Data (ICDD).

Tests description

Effect of water quality on silica removal. Having found the best conditions of pH and velocity gradients in the reactor and the flocculator (Villegas-Mendoza, 2011), data not shown in this paper, continuous runs were performed for each type of water using the EC small pilot. The values of pH that optimize aluminum flocks formation and consequently silica removal were as follows: RW1 = 7, RW2 = 6, CTBW1 = 5.5, and CTBW2 = 5, it was noticed that as sulphate concentration increases pH value should decrease in order to achieve the highest aluminium hydroxide precipitation.

The aluminium dose was set at ~60 mgl^{-1}, which corresponds to the highest concentration that can be obtained with the power source used. This was done in order to analyze the efficiency of high doses of aluminium to remove silica. The response variables were measured every one or two hours along the duration of the tests, which provided between 6 and 8 operation data in a steady state system. This allowed to obtain enough data to carry out a statistical analysis applying a Student´s test (comparison of two means), and to compare the results among the different kinds of water.

As the head loss in the EC reactor began to increase (figure 3b), the flow was increased in order to drag hydraulically deposits that form on the electrode surface, except in the case of

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CTBW2, that was used as reference to see what is the result when this operating condition was not implemented.

The operation of the system was stopped when the voltage began to increase. The increasing in the voltage is considered as an indicative of the electrodes passivation. The increase in head loss suggests mechanical obstruction in the reactor.

Cost of treatment. Once the behavior of the four types of water was analyzed, the energy costs for the oxidation of aluminium, as well as those of aluminium foil itself and chemicals needed to set pH at the optimum values of operation, were calculated for each matrix.

These factors were considered because they are the main contributions and can widely vary depending on the water quality characteristics. The cost for aluminium oxidation depends on voltage, which is a function of the conductivity. The required doses of aluminium depend on the silica concentration and the amount of acid necessary to reach the pH to maximize the silica removal depends on characteristics of the water such as alkalinity.

The following data were considered for cost calculation: a) electric energy = US$0.12 KW-h; b) aluminium plates = US $4.85 / kg aluminium; c) Hydrochloric acid to adjust pH = US $0.16 / L; at a rate exchange of $14.00 MXN = 1 US $.

Results and discussion

Analysis of the effect of water quality

The four types of water were tested using continuous EC small pilot system between eight and ten hours per day and the data were plotted with respect to real hours of work. Figure 3a shows the voltage with respect to time of operation. This voltage is the necessary supplied voltage to guarantee the required electric current for the aluminium production pursued. Two observations can be done according to the tendency of this variable: the voltage with clean electrodes depends inversely on the conductivity, and the passivation starts in general after approximately 10 hours of work. The passivation is a phenomenon characterized by the gradual increase in the electrical power required to set the desired current in the electrolytic cell during the operation of the system. In this case, the passivation does not depend significantly on the quality of the water, but may depend on the current density, that was a fixed parameter similar for the four types of water.

Figure 3b shows the hydraulic head loss, measured with the mercury manometer, with respect to time of operation. In all the tests the head loss increased significantly between the fourth and sixth hour of operation. Once the
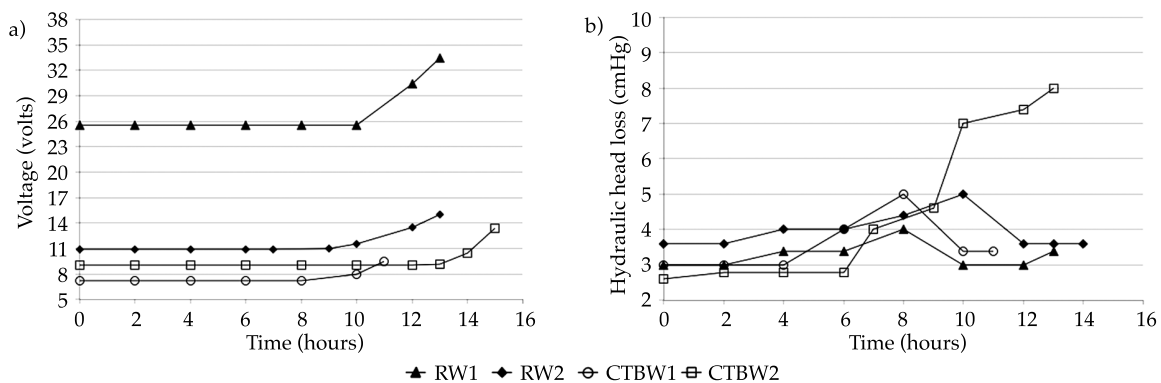

Figure 3. Variation of a) voltage, and b) hydraulic head loss in relation to operation times.

head loss increased, we look to push it down by hydraulic flushing, increasing drastically the flow of operation for ~ 10 seconds (except in the CTBW2 test, where this washing method was not implemented as a measure of comparison). This allowed a temporary recover of the initial head loss and the cleaning off of most of the soft and spongy deposits present on the electrodes, however, it did not help to stop the passivation process, indicating that this phenomenon is associated with the presence of deposits strongly adhered to the electrode surface. Through studies of powder X-ray diffraction, the main component of the inlay was identified as bayerite insoluble $Al(OH)_3$ when we used well water in the tests.

This second type of deposit can be partially removed with chemical washing and can be completely removed only by mechanical abrasion.

It has been consistently observed during the here presented and previous experiments, that an increase in the head loss is always followed by an augment in the voltage. This is important because once the head loss increases we can expect that the passivation of the electrodes shortly begin.

In figure 4, the concentrations of silica and aluminium at the filter exit are shown as a

function of time. Only data from the second hour and beyond have been considered for the statistical analysis, because the hydraulic residence time for the treated water in the system is around 2 h. As can be observed in figure 4b, due to the optimization performed with the settler-filter looking for concentrations below 0.2 mgl^{-1}, the aluminium was almost totally removed in every case, corresponding to a removal efficiency of more than 99%.

From the Gelover-Santiago *et al.*, study (2012), the values for the relation mgl^{-1} dosed Al^{3+}/mgl^{-1} of removed SiO_2, measured at the exit of the settler, were in the interval 1.22 ± 0.12 while for the present job were 1.36 ± 0.05. According to the t-test, both media values belong to the same interval at a 95% level of confidence. This is an indication of the reproducibility of the system.

The amount of residual dissolved silica and the relation dosed Al^{3+}/SiO_2 removed have more reproducible values along the experimentation time for tests with make up water (groundwater) than those observed in cooling tower blowdown (figure 4a). In the same figure it can be observed that more silica was removed from blowdown water than from groundwater.

The statistical analysis showed that the silica removed in both blowdown samples had
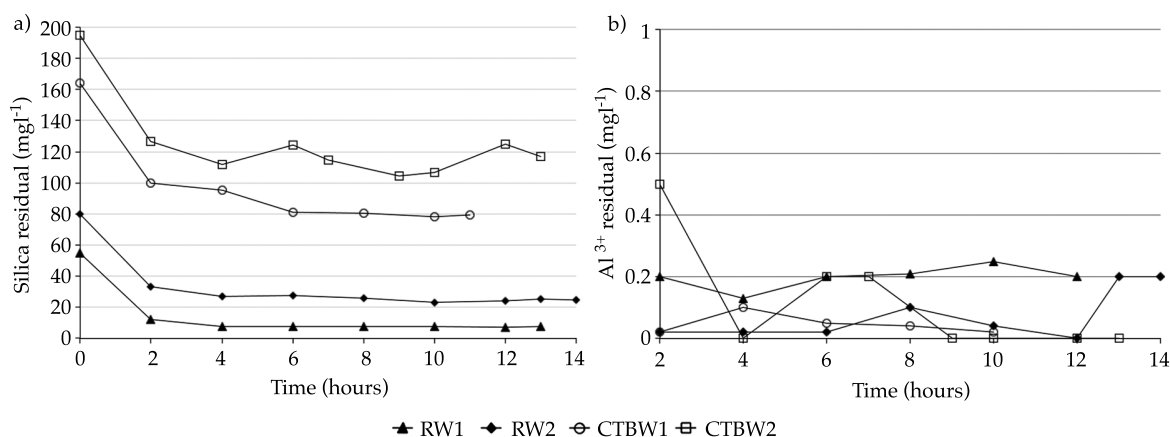

Figure 4. Variation of a) residual silica, and b) residual Al^{3+}, in the filter effluent, with respect to time of operation.

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not significant differences (95% of confidence), but, a different situation occurs for the silica removed in the two groundwater samples, where the removed silica from RW1 was different from that from RW2. In addition, the amounts of removed silica were statistically different between groundwater and blowdown water (figure 5).

This behavior has nothing to do with the aluminum produced, because the means of this parameter are significantly equal between the four types of water studied, contrary to what would be expected based on their chloride concentration, because as reported in the literature (Vargel, 2004), this ion favors the oxidation of aluminum and therefore its production. However, no clear relationship was observed with respect to this ion. Other authors report on the combinated effect of the presence of sulphate and chloride ions. In our case the four matrices contained a certain amount of sulphates, according to Trompette and Vergnes, 2008, and Huang *et al.* (2009), sulphates can preserve the passive alumina film (Al_2O_3) naturally present over aluminium surface, situation that can counter the corrosive effect of chloride ions.

The greater removal of silica observed with the two types of blowdown water, with respect to the two types of well water, is rather explained by the fact that the sedimentation step is more efficient when the water contains

suspended solids, as is the case of purge water. However, the higher efficiency in RW2 with respect to RW1 can only be explained by a higher conductivity, or that RW2 starts with a higher concentration of silica. This behavior requires further study, but has been observed by our working group, that the lower the concentration of silica, the system is less efficient.

The dissolved silica after sedimentation and after filtration were almost the same and something similar occurred with the final concentrations of aluminium, this fact means that the sedimentation step was highly efficient in removing the flocks, and that filtration is a polishing step.

The analysis of the relation dosed Al^{3+}/ SiO_2 removed confirmed that silica removal is more efficient for blowdown water than for groundwater. The amount of aluminium necessary to remove one unit of silica is higher than the unit in the case of groundwater while is around 0.8 for blowdown water. The complexity of the blowdown water matrices, rich in salts and with the presence of diverse chemicals such as dispersants, biocides and corrosion inhibitors is an important factor that must be studied separately to identify its impact in the proposed process.

Treatment cost

After quantify the cost of the treatment, where the amount of aluminium produced, the acid consumed and the electric energy were considered, it was clear that water conductivity is a very important parameter to be consider as it strongly affects the final price of the treatment (figure 6). In the present study RW1 (US $0.74m^{-3}$) had the highest treatment cost due to its low conductivity which significantly increases electrical energy consumption. The costs for the other three types of water were: RW2 (US $0.52 m^{-3}$), CTBW1 (US $0.53 m^{-3}$) and CTBW2 (US $0.52 m^{-3}$), i.e. almost the same price for similar aluminium doses. It must be known that the unitary prices will be higher
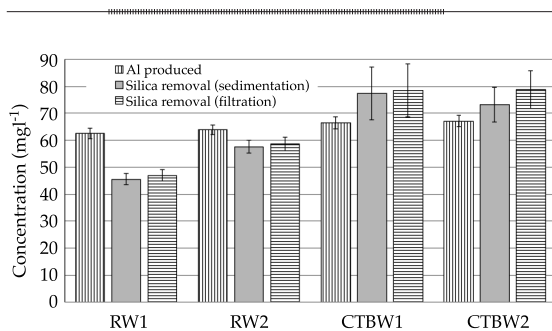

Figure 5. Produced aluminium and removed silica. Average values and confidence intervals are shown in each case for every one of the four types of water studied.

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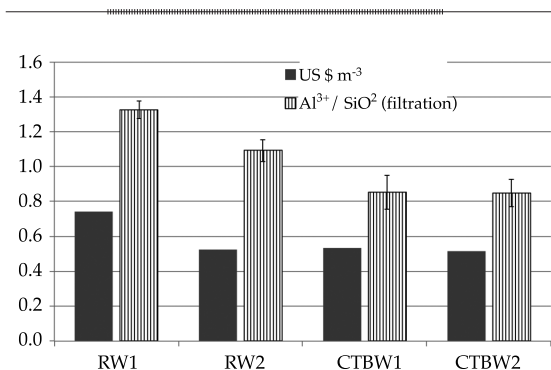
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Figure 6. Relation produced aluminium-removed silica and unitary cost per cubic meter.

for higher concentrations of silica, as it will demand higher doses of aluminium per cubic meter of treated water and then more electric current.

Cost for CTBW Treatment

Besides the cost and efficiency of the treatment, another factor to be considered in deciding the matrix to treat, is the desired final concentration of silica and the possibilities for reuse the recovered water. In the present study, RW2 is the replacement water used in a cooling tower whose blowdown current is CTBW1, this CT works at CC = 1.97 (164 mgL s^{-1} of silica in blowdown and 83 mgL s^{-1} of silica in the make up). Applying the proposed treatment to CTBW1 allowed us to reach a silica concentration similar to the one present in the make up water source. In this situation it is possible to recover all the water from the blowdown and reuse it in the system as make up water managing the same CC, saving first use water and with no blowdown discharge.

As a manner of illustration if E = 100 L s^{-1}, RW2 = 203 L s^{-1}, and CTBW1 =103 L s^{-1}, according to equations 2 and 3. Treating CTBW1 has a cost of US $ 0.53 m^{-3}, the convenience or not of treating such a current depends on the price of groundwater; i.e. on what is more expensive extracting groundwater or treating the blowdown.

In contrast, if one decides to treat the RW2 current instead of CTBW1, the initial concentration of silica can be lowered from 83 up to ~25 mg/L making then possible to reach up to 6.4 CC. In this case, using equations 2 and 3 for the hypothetic situation of the previous paragraph, it will be necessary to treat 118.5 L s^{-1} of make up water at a cost of US $ 0.52 m^{-3} with a rate of 18.5 L s^{-1} blowdown. It means that for this particular example the cheaper option is to treat the blowdown current.

In a second case, where the CT blowdown had the CTBW2 characteristics, and the hypothetic requirement to be reused as a make up water was to reach a final silica content of 50 mgl^{-1}, the treatment would not allow such condition, so the cost to down the silica concentration from 195 mgl^{-1} to 50 mgl^{-1}, will be very high. Under these conditions the dilemma is whether to treat the blowdown or continue using groundwater as make up. The final decision will depend on the water availability and price.

Conclusions

Electrocoagulation is a technically feasible option to reduce silica concentration in cooling towers water. The application of this treatment could help to reduce the huge amount of make up water usually demanded by cooling processes.

The results obtained in this job allow us to conclude that the cost of the treatment is an inverse function of water conductivity, however the function is not linear and for the conductivities explored in this job, higher than that for the RW1 water, there were not significantly differences in the energy consumption associated with the conductivity.

The amount of silica to be removed is another important parameter to be considered in the cost as it determines the amount of aluminium to be dosed. There is a linear function between the amount of aluminium and the required electric current (energy). For this parameter the relationship dosed aluminium- required energy is linear for a regime of constant flux.

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The overproduction of aluminium must be strictly controlled otherwise the relation dosed Al^{3+}/SiO_2 removed increases significantly just making the treatment more expensive without any additional benefits.

The stoichiometric relationship found in this job between aluminium and silica is nearly 1:1 (mass:mass). This value is significantly less than that reported in the literature for the conventional application of aluminium salts where the relation is 4:1 aluminium:silica.

For the studied conditions the proposed treatment was more efficient in terms of the relation dosed Al^{3+}/SiO_2 removed when it was applied to blowdown water and it was associated with the higher content of suspended solids for this matrix, even though this fact requires additional studies. However to work with blowdowm water causes a major variability in the system and since the water quality of this matrix can be very variable along a daily basis, the control of the treatment system turns complicated.

The decision of which of the matrices to treat, make up water or blowdown, in order to increase the concentration cycles to save water is strongly dependent on the silica concentration of both matrices, on the make up water quality and on the water availability and price. Treating blowdowns could be convenient for high water scarcity zones.

The process will result attractive for systems with a low value of CC. In the case of actually high CC, the relation silica in blowdown/silica in make up water is expected to be high and as we have shown, the removal of a high silica concentration will be very expensive without any additional real benefit.

Acknowledgements

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