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Innovative geocomposite for dredged sediments depollution

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Abstract. In France, sediments from river dredging are considered as waste. Without any special treatment, they must be stored in landfills. The dredged sediments of our area of study (North of France) are polluted with trace metals (antimony and zinc are above the French regulation threshold). The main objective of this study is to develop a new method of active depollution and dewatering treatment in order to enable the reuse of treated sediments. This treatment is carried out through ponds equipped with innovative geocomposite. This innovative geocomposite has two typical passive functions (filtration and drainage). In addition, it is functionalized by an innovative electrokinetic technology to confer it active roles (depollution and dewatering). Electrokinetic consists in applying a difference of electrical potential to a porous medium. We present here the results of a preliminary laboratory study that simulates this new method on a short-term (3 weeks) experiment. The results show that the electrokinetic treatment has an impact on the chemical and physical properties of sediments. The treatment reduces by 50% the concentration of zinc in the leachate after lixiviation.

1. Introduction

Today, the issue of sustainable development is topical. Thus, several solutions have been developed in different fields such as energy with green energies and construction, with more ecological materials. In the same perspective of practicing a more sustainable management of waterways, a new method of sediment treatment resulting from river dredging is being developed. In fact, in France, sediments from river dredging are considered as waste as soon as shore management is considered. Without any special treatment, the polluted sediment must be stored in landfills. The dredged sediments of our area of study (North of France) are polluted with trace metals (antimony and zinc are above the threshold from the French regulation). These waste storage facilities require more and more space and alter the ecological balance of the areas where they are located. In addition, to avoid groundwater pollution, the landfills must be lined which increases the cost of managing these sediments. The main objective of this study is to develop a new method of active depollution treatment in order to enable the reuse of treated sediments. In France, the inert nature of such sediments is defined by Article R.541-8 of the French Environmental Code [1] as any waste that does not undergo any significant physical, chemical or biological modification and does not deteriorate the materials with which it comes into contact in a manner likely to result in harm to the environment or human health. The thresholds taken into account in this regulation to determine the inertness or not of the sediments are those of the admission thresholds in a landfill defined by the European decision of 19 December 2002 [2] concerning the standards for admission to landfills. These thresholds are measured by a leaching test according to standard NF EN 12457-2 [3].



This treatment is carried out through ponds equipped with innovative geocomposite. The innovative geocomposite has two typical passive functions that are filtration and drainage. In addition, the geocomposite is functionalized by an innovative electrokinetic technology to confer it active roles. Electrokinetic consists in applying a difference of electrical potential to a porous medium. Electromigration and electroosmosis are two of the phenomena that confer an active depollution and dewatering roles to the innovative geocomposites. Electroosmosis is a flow of fluid in the matrix of charged particles that appears when an electrical potential difference is applied. The application of an electrical potential difference in a porous medium causes the appearance of an electroosmotic flow due to the displacement of the charges bound to the surface of the particles. Electroosmosis allows the decontamination of sediments by the transport of solubilized pollutants out of sediments and intervenes for the extraction of sediments by the mobilization of interstitial fluid. Electromigration or migration of ions is the movement of ions due to the application of an electrical potential difference. During the application of an electrical potential difference, the electrodes are polarized. The anode is the positive electrode and the cathode is the negative electrode. Under the effect of the electrical potential gradient, cations (positive sign ions) migrate to the cathode and the anions (negative sign ions) migrate to the anode. Pollutants must be solubilized first to be mobilized by electroosmosis and electromigration [4]. Indeed, during the application of electrokinetic we observe the appearance of a basic front at the cathode and an acidic front at the anode. The acidic front is beneficial for the electrokinetic treatment because it allows a solubilisation of trace metals, but the basic front is detrimental because it causes the precipitation of some trace metals. This heterogeneous spatial distribution of pH causes other heterogeneities such as the development of a heterogeneous flow through the electroosmotic permeability coefficient that is sensitive to pH, and the development of a heterogeneous interstitial pressure that can lead to the appearance of suction near the cathode [5]. Despite the complex phenomena occurring during the treatment, electrokinetic has been used successfully for depollution and dewatering. Indeed, Bourges-Gastaud *et al.* [6] developed an experimental device to test the ability of an electrokinetic geocomposite to dewater mature mine tailings. Their tests showed that the electrokinetic combined with the mechanical dewatering extraction made it possible to increase the solid content of the mine tailing from 42% to 66% and to increase the shear strength from 0 to 40 kPa on average. Wang *et al.* [7] managed to reduce by 43% (6 days of treatment and a current density of 0.390 mA/cm²) the concentration of arsenic. This explains why we chose to study electrokinetic for sediment treatment.

The innovative geocomposite is functionalized with the aim of incorporating it an active depollution role and an active dewatering role. In the configuration of ponds, the electrodes are set up horizontally to respect the primary objective of the ponds, which is the gravitational dewatering (see Figure 1). The flow produced by the electrokinetic is in the direction of the gravitational flow and a constant water supply is provided at the surface of the pond (at the anode) to allow a permanent flow during the treatment. The flow crosses the sediments in order to solubilize the pollutants and in order to extract them.

This paper presents the experimental study of the sediment treatment of a dredged material by this innovative geocomposite. An experimental cell has been developed to simulate in laboratory the treatment conditions in ponds.

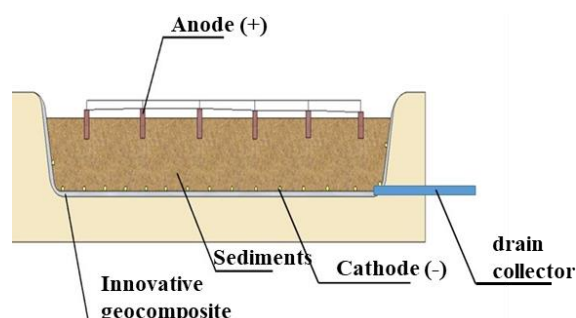


Figure 1. Principle of the treatment pond

2. Material and method

2.1. Material

2.1.1. *Sediments*. The sediments come from a storage pond belonging to "Voies navigables de France (VNF)". This pond is located in the city of Noyelle-sous-Lens (North of France). The concentrations of the chemical elements present in the sediments are presented in Table 1.

Table 1. Chemical composition of the studied dredged sediment; concentrations of total trace elements and after leaching (D.M: dry matter)

Metal	Unit	Initial	Inert threshold (following [2])
Arsenic (As) ^a	mg/kg D.M.	16.9	
Cadmium (Cd) ^a	mg/kg D.M.	6.1	
Chrome (Cr) ^a	mg/kg D.M.	61.1	
Copper(Cu) ^a	mg/kg D.M.	315	
Lead (Pb) ^a	mg/kg D.M.	246	
Mercury (Hg) ^a	mg/kg D.M.	1.16	
Molybdenum (Mo) ^a	mg/kg D.M.	2.02	
Nickel (Ni) ^a	mg/kg D.M.	31.2	
Zinc (Zn) ^a	mg/kg D.M.	1100	
AFTER LEACHING TEST (NF EN 12457-2)[3]			
Antimony (Sb) ^b	mg/kg D.M.	0.078	0.06
As ^c	mg/kg D.M.	<0.2	0.5
Barium (Ba) ^c	mg/kg D.M.	0.31	20
Cd ^b	mg/kg D.M.	0.033	0.04
Cr ^c	mg/kg D.M.	<0.1	0.5
Cu ^c	mg/kg D.M.	0.28	2
Pb ^c	mg/kg D.M.	0.17	0.5
Hg ^b	mg/kg D.M.	<0.001	0.01
Mo ^b	mg/kg D.M.	0.241	0.5
Ni ^c	mg/kg D.M.	0.15	0.4
Selenium (Se) ^b	mg/kg D.M.	0.031	0.1
Zn ^c	mg/kg D.M.	4.02	4

^a NF EN ISO 11885[8] – NF EN 13346 Method B[9]

^b NF EN ISO 17294-2 [10]/ NF EN 16192[11]

^c NF EN ISO 11885[8] / NF EN 16192[11]

2.1.1. Experimental cell. A new experimental cell was developed to conduct this experiment. The schematic diagram of the experiment is given in Figure 2. The experimental cell has the following dimensions: 0.25 m wide by 0.35 m long and 0.60 m high. It has orifices for inserting electro conductivity (EC) probes. The bottom of the cell is lined up with a drainage geocomposite (filtering and draining nonwoven needle-punched geotextile with mini-drain), functionalized by an electrode to decontaminate the sediment. The surface of the sediment is lined with an electrode (the anode) in titanium alloy coated with mixed oxide of precious metals to enable it to be corrosion-proof. The anode has a shape of a grid. The experimental cell is equipped with the following sensors:

- EC probes follow the evolution of the electrical conductivity inside the sample. These probes are placed at three points: one in the anode area, one in the cathode area and one in the central area of the sample;
- A LabVIEW control panel and an electrical resistance of 3 Ohms follow the evolution of the intensity during the treatment;
- A scale measures continuously the amount of water used for the depollution and the amount of water extracted during the dewatering phase.

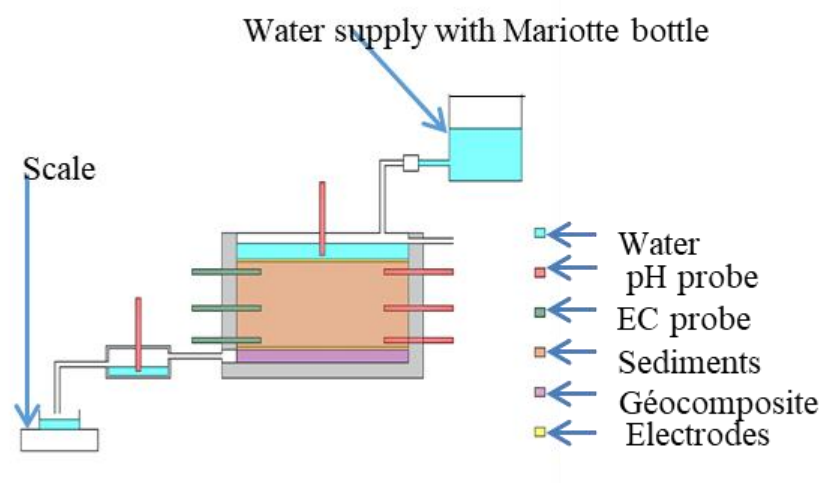


Figure 2. The experimental cell principle

2.1.2. Innovative geocomposite. The innovative geocomposite is a drainage geocomposite. This drainage geocomposite has intrinsic functions which are:

- Filtration, which is the retention of soil or other particles subjected to hydrodynamic forces while allowing the passage of fluids through or in a geosynthetic material [12]. In the geocomposite the filtration function is provided by a geotextile filter. The suitability of geotextile for sludge filtration has been proven and filtration criteria defined [13-17]. The geotextile filter only acts in the first moments of the filtration to allow the formation of the "filter cake", which then ensures the role of filter. The geotextile filter follows the recommendations of Bourgès-Gastaud *et al.* [17] and Delmas *et al.* [13];
- Drainage, which is the collection and transport of rainwater, groundwater and / or other fluids in the plane of a geosynthetic [12]. The drainage function is provided by a needle-punched nonwoven drainage core and mini-drains.

In addition to its intrinsic functions, the geocomposite is functionalized by the electrokinetic technology to give it active roles of depollution and dewatering. The geocomposite is functionalized by the addition of a galvanized steel electrode in 35 mm² wire form. The wire-shaped electrode is placed in the same gusset as the mini-drain (see Figure 3). The characteristics of the geocomposite are shown in Table 2.

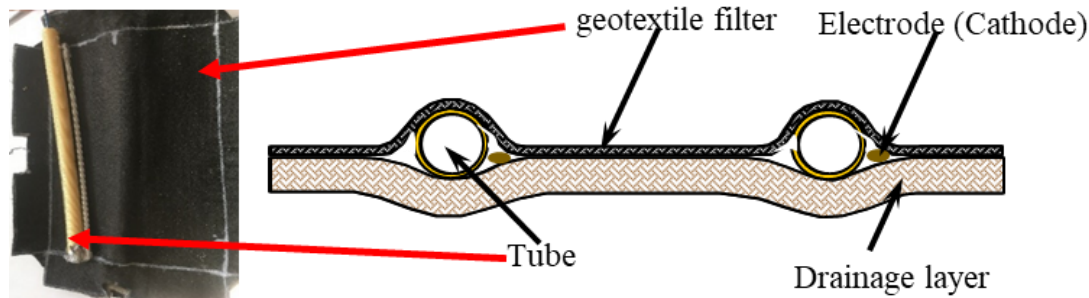


Figure 3. The innovative geocomposite material. At the left, photography of top face of innovative geocomposite (geotextile filter in black) and tube with wire electrode. At the right, schematic representation of the section in the thickness of innovative geocomposite

Table 2. Characteristics of the geocomposite

Properties	Values	Standard Methods
Thickness under 2 kPa normal load	5 mm	NF EN ISO 9863-1[18]
Filtration opening size	110 μm	NF EN ISO 12956[19]

2.2. Method

2.2.1. *Depollution experiment.* The sediments (60 kg with initial height of 44 cm) are introduced into the cell with an initial solid content (SC see equation (1)) of 50% in successive layers.

$$SC = \frac{\text{solid mass}}{\text{solid mass} + \text{liquid mass}} \times 100 \% \quad (1)$$

Beforehand, the sediments are sieved through a 6 mm sieve to remove coarse particles, and re-wetted with distilled water to a solid content of 50%. The valve allowing the supply of water is opened in order to maintain a constant height of water. It is necessary to keep a constant water level at the anode in order to facilitate the electromigration phenomenon, and to allow electrokinetic continuity over time. Dewatering of the anode could even result in stopping of electrokinetic. The constant water level is maintained for 48 hours to measure the hydraulic conductivity of the sediments (Phase 1). Then the electrokinetic is started with constant water supply (Phase 2). At the end of Phase 2, the electrokinetic is maintained and the constant water supply is stopped. The sediment is dewatered by electrokinetic until electrokinetic stop (no more water extracted at the cathode) (Phase 3). At the end of the dewatering, the cell is disassembled. The final pH and the final mass water content are measured on the thickness of the sediment column. Then the sediment is divided into three equal areas: the anodic area, the central area and the cathodic area and the pollutant concentration are measured in these areas.

2.2.2. *Chemical analyses.* Different chemical analyses are carried out on the three areas of the sediment:

- On the solid part (sediments): total concentration of trace metals. This analysis is performed after acid digestion (regal water mineralization - heating block) according to standard NF EN ISO 11885[8] – NF EN 13346 Method B[9];
- On the eluate after a leaching test of the sediment performed according to standard NF EN 12457-2 [3]: total concentration of trace metals in eluate after filtration. The leaching test is

used to determine the sediment's ability to release trace metals under the effect of a stream of water.

3. Results

The results presented in this section were obtained for an experiment running over a period of 3 weeks. This period is relatively short compared to the height of the sediments column to be treated (i.e. 44 cm).

3.1. Final state of innovative geocomposite after treatment

After the treatment, the innovative geocomposite was disassembled and weighed. The innovative geocomposite was in a good visual state, with impregnation of sediment and precipitate (see Figure 4). Table 3 shows the state of impregnation of the innovative geocomposite and conductive materials. Conductive materials did not lose mass during processing and are resistant to corrosion. The cathode has a mass gain due to the incorporation of precipitate within it.

Table 3. Variation of mass in the geocomposite and conductive material at T0 and after the 3-weeks experiment

	Initial mass (g)	Final mass (g)	Impregnation rate
Filter+ drainage layer	84.11	164.2	542.7 g/m ²
Mini-drain	33.12	34.15	2.94 g/m
Anode	44.26	44.26	
Cathode	106.2	107.3	

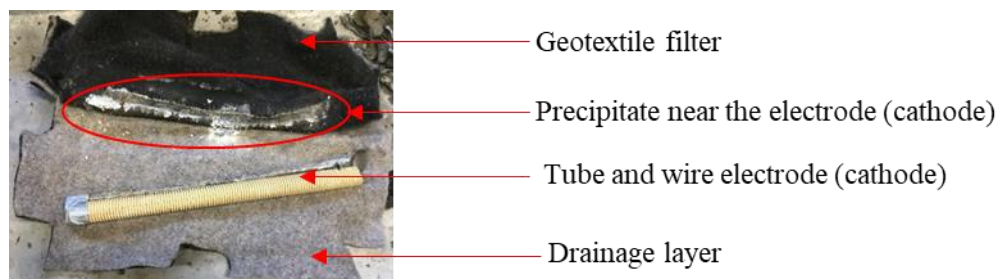


Figure 4. Final state of disassembled innovative geocomposite after the 3-weeks experiment

3.2. Physical parameters

The following caption is used in all section 3.2. and allows to follow the duration of each phase of treatment:

- | (black line): Starting electrokinetic
- | (green line): Stopping the water supply
- | (brown line): No more supernatant at the anode
- | (red line): Stopping electrokinetic

3.2.1. *Evolution of the pH.* Figure 5 (a) shows the evolution of the pH level of the liquid extracted at the cathode, the supernatant at the level of the anode and the middle of the sediment. The pH level of the fluid extracted at the anode remains stable during the first 20 hours of the electrokinetic treatment before starting to grow rapidly to reach a value of 12 and remains stable during the treatment. The pH in the middle of the sediment remains the same throughout the treatment with a value of about 6.8. The pH sensor at the anode supernatant failed, the pH was measured punctually and is around 1.8. Unfortunately we do not have the kinetics of pH evolution at the anode supernatant level. Figure 5 (b)

shows the evolution of the pH on the thickness at the end of the experiment. There is a heterogeneity in the thickness. The pH rises from the anode to the cathode from 6 to 11.5 with a sharp growth at 5 cm from the cathode. Indeed at 7.5 cm from the cathode the pH is about 6.9 while at 2.5 cm it is 11.5. This heterogeneity is not only observed on the pH but also on the electrical conductivity, as we will see in the section 3.2.5.

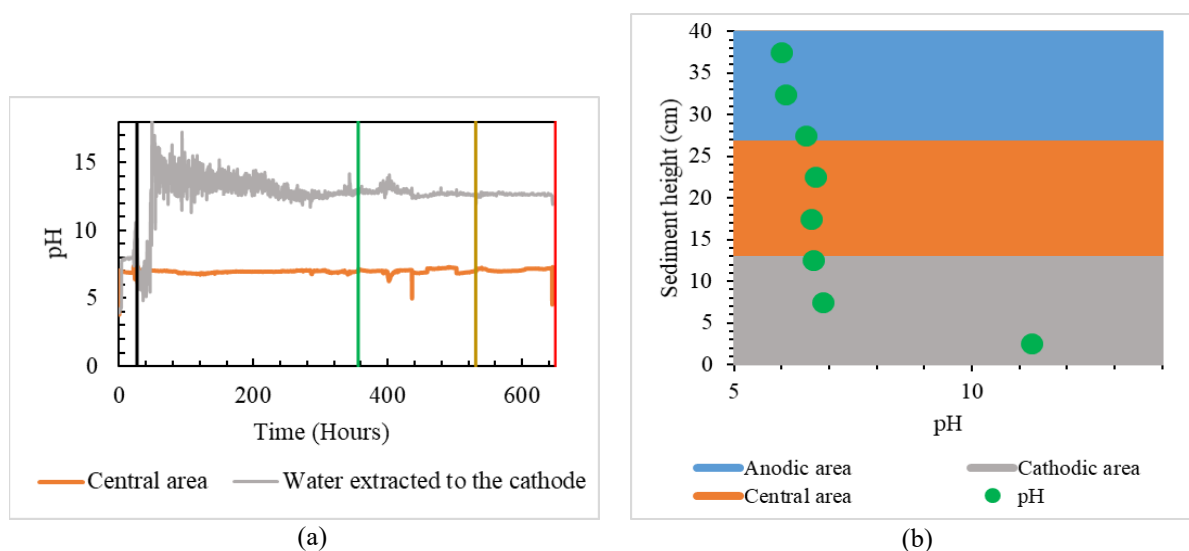


Figure 5. (a) Evolution of pH as a function of time during the treatment within the cell (b) Spatial profile of pH along the height of the sediment (at the end of experiment).

3.2.2. Settlement. Figure 6 shows the evolution of the height of the sediment during the electrokinetic treatment. The settlement occurs from the first moments and especially during the period of gravity dewatering. Indeed, during the gravity period, the sediment height decreases by 2.5 cm and then the kinetics of settlement slows down. During the first moments of the electrokinetic treatment (phase 2), the sediment height decreases by 1.5 cm and stabilizes at 41.5 cm and remains unchanged throughout the treatment. The settlement during the electrokinetic treatment occurs at the same time as the appearance of the basic pH at the cathode which causes a suction in the cathodic area and therefore a consolidation which could explain this settlement.

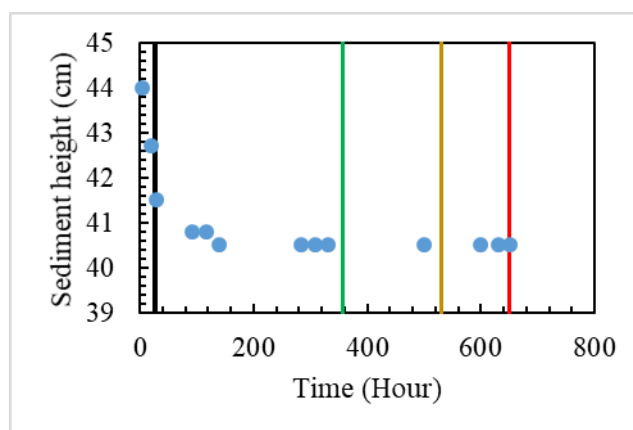


Figure 6. Evolution of the height of sediment in the experimental cell as a function of time during the 3-weeks treatment

3.2.3. Mass of water extracted at the cathode. The mass of water extracted increases almost linearly during the first two phases (see Figure 7 (a)). This trend has been observed by several authors during depollution treatment [20-23]. In the last two phases of the treatment the increase in the mass of extracted water decreases due to the stoppage of constant supply of water which causes a decrease in the hydraulic load. The mass of water extracted reaches a plateau and stagnates during the last phase because there is no more supernatant. By observing Figure 7 (b), the flow rate decreases during phase 1. It can be due to the settlement that the sediment undergoes, which modifies the state of consolidation of the latter and consequently its hydraulic conductivity. During the second phase, the application of electrokinetic increases the flow rate due to the addition of the electroosmotic flow and the hydraulic flow. Then the flow decreases progressively, which may be due to the heterogeneity of pH which leads to a heterogeneous electroosmotic flow by modifying the coefficient of electroosmotic permeability.

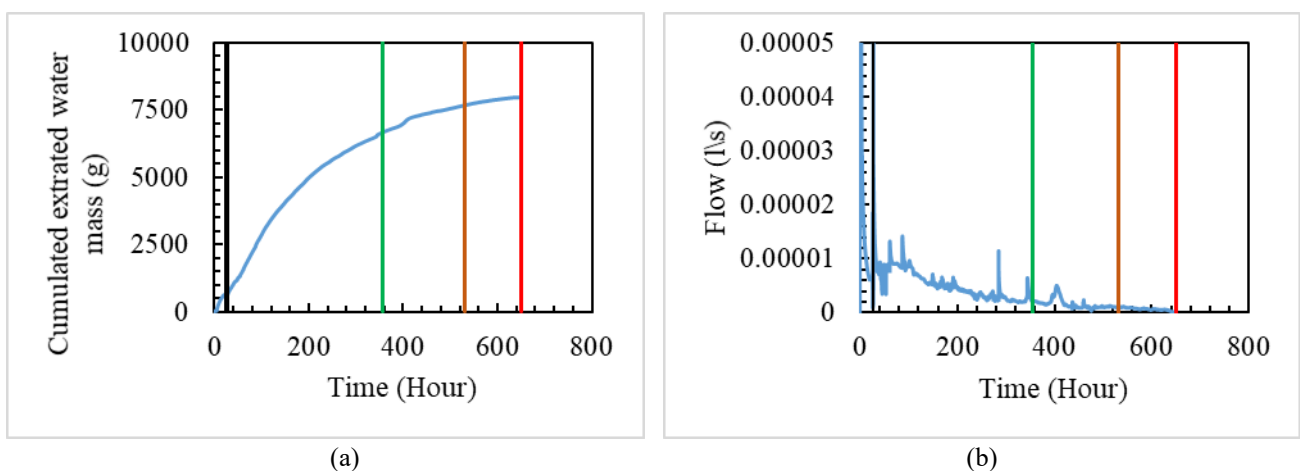


Figure 7. (a) Evolution of cumulated extracted water to the cathode as a function of time.
(b) Evolution of flow as a function of time

3.2.4. Electrical intensity. Figure 8 shows the evolution of electrical intensity as a function of time. The intensity is at its maximum as soon as the electrokinetic are installed and reaches a value of 450 mA. Then the electric intensity gradually decreases to reach a minimum value around 50 mA. This behaviour has been observed in several depollution or dewatering studies [23-25]. At the beginning of the treatment, the ions are present which explains the high value of the intensity. During the treatment, the ions concentration decreases due to electromigration, absorption and precipitation reactions. This drop in concentration causes a decrease in electrical conductivity and this explains the decrease in intensity during treatment. The basic medium at the anode causes precipitation of trace metals that decreases the conductivity.

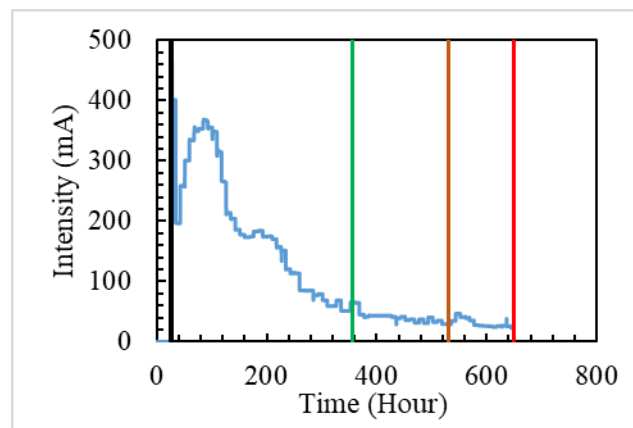


Figure 8. Evolution of electrical intensity during the treatment as a function of time.

3.2.5. Electrical conductivity. In Figure 9 (a), a single value of electrical conductivity is observed throughout the medium in the initial state. Over time, the electrical conductivity of the sediment is reduced at the cathode area. Firstly, this can be explained by the fact that the cathodic area is gradually settled by the weight of the sediments. Secondly, because of the appearance of the basic front, the ions precipitate at the cathode area, which causes a decrease of the electrical conductivity of the medium. For the anode and the middle area, an increase of electrical conductivity is observed at the beginning of the treatment followed by a decrease. The tendency of the electrical conductivity of the sediment at the anode is similar to that of the electrical intensity. The electrical conductivity of the medium is experiencing a sudden drop which appears 40 hours after the start of electrokinetic. This sudden drop is due to the appearance of the basic pH at the cathode that causes suction in the cathode area, a settlement and therefore a decrease in porosity. The electrical conductivity of the liquid extracted at the cathode gradually increases from the start of the electrokinetic to a maximum value of 4 dS/m and remains around this value throughout the treatment (see Figure 9 (b)). This denotes the migration of ions out of the sediment due to electrokinetic and the presence of ions in the liquid extracted at the cathode increases its electrical conductivity. The sensor at the anode level has failed so we cannot interpret the results of this sensor.

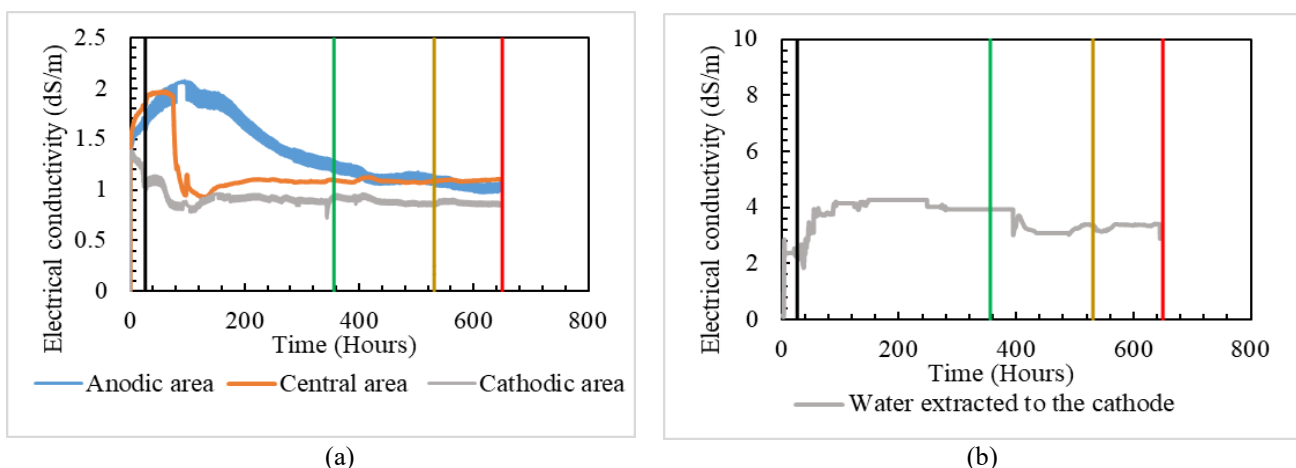


Figure 9. (a) Evolution of electrical conductivity in the sediment as a function of time during treatment. (b) Evolution of electrical conductivity in water as a function of time during treatment.

3.2.6. *Water content.* Figure 10 shows the evolution of the mass water content in the sediment. The final mass water content in the sediment decreases from the anode to the cathode which is in the opposite direction of the flow from the anode to the cathode. Other studies have observed the same pattern of mass water content [26-28]. This behaviour can be explained by the heterogeneity that develops in sediments during electrokinetic treatment.

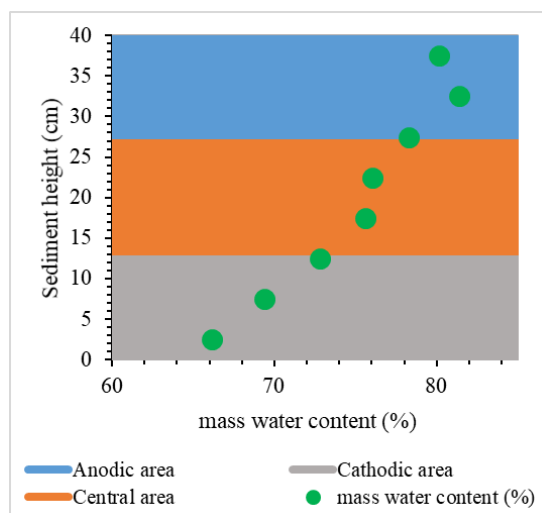


Figure 10. Spatial profile of the mass water content along the height of sediment at the end of the 3-weeks experiment.

3.3. Chemical parameters

In this part we present the results of the chemical analyses. The analyses are made over three specimens taken from the anodic area, the cathode area and the middle of the sample. The comparisons with the initial values are indicative because we did not have the opportunity to have an initial pollutant content of the sediments used for the experiment. Therefore we used the values measured at the sampling site as initial pollutant content values.

3.3.1. *Analyses on sediment.* Figure 11 shows the normalised concentration of trace metals in the sediment after treatment in the different areas. Cd, Cr, Pb and Zn normalized concentration is equal or less than unity for anodic and central area and superior to unity for cathodic area. It means that the concentrations reduce at the anodic and central areas and increases at cathodic area. For As, Hg and Ni the normalised concentration has opposite trend. The difference of normalised concentration between the different areas is small that could be explained by the short treatment time and the low refinement of the thicknesses of the concentration measurement area. From these observations two behaviours emerge :

- the first is a cationic behaviour, that is to say that the metals are present in a cationic form and move from the anode to the cathode under the effect of electromigration and electroosmosis. These metals are cadmium, chromium, copper, lead and zinc; Concentrations increased at the cathode in comparison to the 2 other areas after the 3-weeks experiment.
- the second is an anionic behaviour, that is to say that the metals are in an anionic form and move from the cathode to the anode under the effect of electromigration but go against the electroosmotic flow. These metals are Hg, As and Ni. Concentrations are slightly higher at the anode in comparison to the 2 other areas after the 3-weeks experiment.

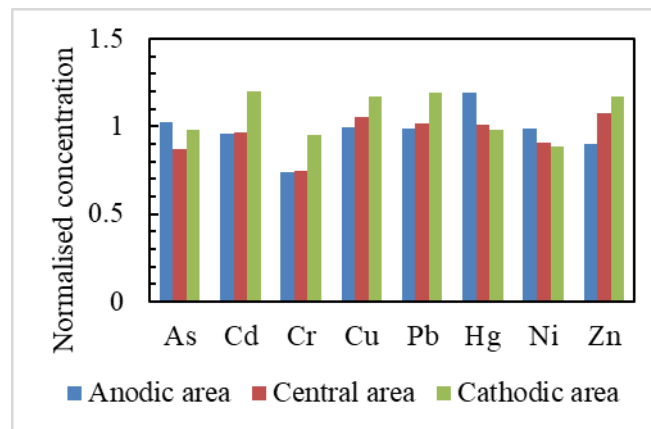


Figure 11. Normalised concentration of trace metals in the sediment after treatment.
 Normalised concentration = $\frac{\text{initial concentration}}{\text{final concentration}}$

3.3.2. *Analyses on leachate.* Figure 12 shows the normalised concentration after standard leaching of the metals in the different areas. We note that some metals such as cadmium have a reverse tendency in leachate in comparison to that of sediments. In fact, Cd has a higher concentration at the anode than at the cathode. This is explained by the fact that Cd precipitates (in a non-soluble form) in the cathodic area because of the basic pH and solubilize in the anodic area because of the acidic pH. Zn and Ni have the same tendency and are below the limit of quantification (of the analytical method) at the cathode area. This implies that electrokinetic treatment can stabilize these metals in forms that do not solubilize so allows sediments to be inert. But other trace metals, like Sb, Ba, Cu, Mo and Se that have a high concentration at the cathode, do not precipitate under the effect of basic pH and thus are not stabilized.

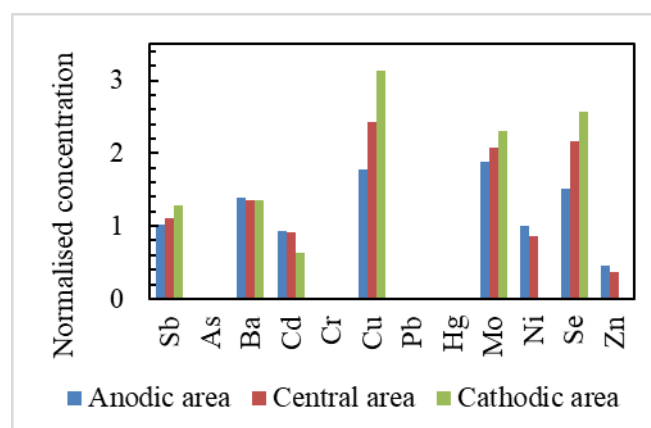


Figure 12. Normalised concentration of trace metals in the leachate after treatment.
 Normalised concentration = $\frac{\text{initial concentration}}{\text{final concentration}}$

4. Conclusions

This paper presents an experimental study of the depollution of dredged sediment from waterways with electrokinetic incorporated on innovative geocomposite. A new experiment cell was developed to simulate the condition of the treatment pond. The first results show that the innovative geocomposite accomplished well its intrinsic functions, the anode is corrosion-proof and the electrokinetic treatment

is very complex and heterogeneous. Indeed during the treatment, there is a modification of the physical properties of the sediment due to the settlement of the sediment, which causes the reduction of the hydraulic conductivity. The application of electrokinetic produces an electroosmotic flow that increases the total flow. This flow decreases during the treatment due to the heterogeneity of pH that induces heterogeneous electroosmotic flow. An acidic condition is developed at the anodic side and a basic condition is developed at the cathodic side. This heterogeneous pH condition is concomitant with a heterogeneous electro conductivity in the sediment. The electro conductivity decreases during the treatment like the electrical intensity. In spite of its complexity, the electrokinetic treatment enables to lower the concentration of zinc in leachates by 50%, and even to eliminate Zn and Ni after precipitation at basic pH near the cathode. In contrast, the concentrations of antimony after leaching does not change at the end of the experiment, so electrokinetic has no effect on Sb, Ba, Cu, Mo and Se. Additional tests over longer periods will be performed to fully evaluate the performance of this new electrokinetic method to depollute dredged sediments.

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