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Potential of Clay-Based Geomaterials for Passive Treatment of Brackish and Saline Waters

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Abstract The increasing salinity and water scarcity of water resources are major problems in semi-arid or arid regions. This study examines the possibilities of a hybrid geomaterial based on clay to treat passively water bodies that are brackish or saline made from locally available Moroccan clays. A quaternary composite was developed by combining ghassoul with kaolinite as well as red ferruginous and illite and green ferruginous illite, to benefit from their complementary physicochemical characteristics. Chemical and mineral analyses reveal the synergistic impact of a high cation exchange capacity as well as iron oxide-driven anionic interaction as well as the stability of the structure. Simulated batch adsorption tests were carried out at beginning NaCl dosage of 1100 mg/L and using different weights of adsorbents. The results demonstrate a gradual reduction in equilibrium salinity as increasing clay dosage, and the maximum capacity for adsorption of 18 mg/g. Adsorption behavior was studied with the help of Langmuir as well as Freundlich isotherm model. While Langmuir analysis yields reliable estimates of the adsorption capacity however, the Freundlich model gives more accurate results, which indicates the multilayer and heterogeneous adsorption that is controlled by several active sites. The results confirm the fact that the composite of hybrid clay permits simultaneous reduction of sodium and chloride ions by combining Ion exchange and surface adsorption processes. With a potential desalination efficiency of 20-30 %, this low-tech solution is a viable and economical alternative for saline water pre-treatment or partial desalination to support ecologically responsible water management strategies in water-poor regions.

Keywords Hybrid clay composite; Passive desalination; Adsorption isotherms; Freundlich model; Langmuir model

1. Introduction

Morocco is battling growing water stress that impacts not only the availability of water, as well as the overall quality of groundwater as well as the quality of coastal water resources[1]. In this regard seawater desalination is now an essential issue that requires the creation of desalination strategies which are economical and sustainable[2]. Desalination methods that are conventional, like reverse osmosis as well as thermal distillation have been extensively used however, they are still associated with significant energy consumption and huge environmental impact, especially those relating to the discharge of brine into coastal and marine ecosystems. To combat these shortcomings the exploitation of clay-based materials using biotechnological techniques has been identified as a promising technology innovation[3]. This strategy makes use of the abundant mineral resources available to allow the transformation of brackish water or seawater to freshwater as well as facilitating the creation of a sustainable, bio-based economy based on bio-based products with high-value-added. In this respect clay-based ceramic membranes are seen as an affordable and ecologically friendly option to desalination, gaining benefits by the use of natural resources in a circular economy model. This is why the focus should be in the sustainable transformation and use of resources, enabling new and innovative technology for water treatment. Clays, Aluminosilicate minerals, are characterised by having a significant specific surface area as well as a substantial cation exchange capacity that makes them very appealing geomaterials for specific adsorption processes[4]. More recent studies[5]., have clearly established the adsorption capacity of clays with Ion exchange mechanisms. The equilibrium of the ion exchange process is typically controlled by a preference towards divalent cations over monovalent ones, based on the mineral's intrinsic properties. The literature emphasizes the electrostatic repulsion of chloride ions from clay layers. This is an important obstacle for their efficient use in desalination processes, and restricts their ability to total salt removal.

2. Intrinsic Properties

2.1 White Clay (Kaolinite)

The clay is a part of the Nador the region of Nador (**Figure 1**) is recognized due to its high silica percentage (SiO_2: 64.14%) and a large amount of Alumina (Al_2O_3: 19.24%)[6]. This mineral's composition is extremely mechanical stability and solid, well-structured as well as a non-swelling, dense porosity. This is the reason kaolinite functions as a solid base material for composites and encourages the physical filtration process through its intrapore-pore diffusion mechanisms[7]. In addition, its non-expansive qualities aid in maintaining structural integrity. It is also one of the main factors in reducing obstruction of pore and fouling issues in clay-based composites.

Fig. 1. Macroscopic appearance of white clay (kaolinite) from the Nador region.

2.2 Red Clay (Ferruginous Illite)

The Rachidia-Guelmima region is the source the red clay (**Figure 2**) is identified by its significant amount of iron oxide (Fe_2O_3: 7.26%) that enhances surface reactivity to anionic species, especially the chloride ions (Cl^-)[8]. In the presence of iron oxide phases results in a pH-dependent point of zero charges (PZC) that plays an important role in the modulation of electrostatic interactions between surfaces. This is a mechanism that counteracts electrostatic repulsion, and also promotes anionic adsorption processes. In turn, ferruginous illite is a significant contributor to the anion-binding capability of the clay-based composite particularly under saline conditions.

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Fig. 2. Red clay (ferruginous illite) collected from the Rachidia–Guelmima region.

2.3 Green Clay (Ferruginous Illite)

It is a product of the Rachidia-Guelmima area green clay (**Figure 3**) displays similar mineralogical properties to red clay. It is however rich by ferrous (Fe2O3: 7.19%) and magnesia (MgO: 3.96%)**[9]**. This particular composition has the highest adsorption as well as detoxification capabilities, which makes it especially effective in capturing dissolving contaminants. It also has an additional porosity, and a higher amount of active surface areas, which aid in anion capture via interactions on the surface. This is why green ferruginous illite is a significant contributor to improving the synergistic performance of the composite of hybrid clay.

Fig. 3. Green clay (ferruginous illite) collected from the Rachidia–Guelmima region.

2.4 Brown Clay (Ghassoul / Stevensite)

It is derived of the Missour region of the Boulemane Province, brown clay (Ghassoul) is primarily comprised of stevensite (**Figure 4**), is distinguished by its significant amount of magnesium (MgO: 15.59%) and lime (CaO: 13.56%). This mineral composition has the highest CEC (CEC) that is due primarily to its layered structure that is expandable. The swelling process assists in the release divalent cations (Mg^{2+} and Ca^{2+}) that can substitute two monovalent sodiumions (Na^{2+}) in saline media[10]. This mechanism of substitution exposes an array of exchange sites that are active, thus improving the efficiency of selective exchange processes. In the end, ghassoul is an essential component in enhancing the effectiveness of removing cations and contributes substantially in the performance overall of the hybrid clay composite brackish and saline water treatment.

Fig. 4. brown clay (Ghassoul/Stevensite) collected from the Missour region, Boulemane Province.

3. Comparative Analysis

3.1 Comparative Analysis of Chemical Compositions

Chemical compositions of four clays shows distinct mineralogical differences that can be explained by their similar behaviour in water treatment saline applications. In **Table 1** the white clay (kaolinite) is heavily dominated by silica (SiO2: 64.14%) and Alumina (Al2O3: 19.24%) which confirms its structural function and its inactivity towards Ion exchange. Contrary to these the red and green clays, which are both ferruginous illites have greater iron oxide levels (Fe2O3 : 7.2 %) that are believed to increase the surface's reactivity and encourage anionic adsorption, especially for chloride ions as is widely reported within the scientific literature[11]. Brown clay (ghassoul/stevensite) is clearly differentiated by its high MgO (15.59%) and CaO (13.56%) contents, reflecting its strong cation exchange capacity and suitability for sodium removal through divalent-monovalent ion substitution mechanisms. The lower concentrations of residual chloride that were measured in the illitic as well as ghassoul clays further demonstrate their ability to be effective in the passive process of desalination. Overall, these variations in composition offer a solid physicochemical base for the synergistic hybrid formulation used during this investigation.

Table 1. Chemical composition of the studied clay materials

Oxide	White (Kaolinite)	Red (Ferruginous Illite)	Green (Ferruginous Illite)	Brown (Ghassoul)
SiO_2 (%)	64.14	63.49	57.53	30.42
Al_2O_3 (%)	19.24	14.31	15.63	1.98
Fe_2O_3 (%)	1.63	7.26	7.19	0.96
MgO (%)	1.53	2.47	3.96	15.59
CaO (%)	2.40	1.72	1.76	13.56
Na_2O (%)	2.98	0.96	0.69	1.13
Chlorides (ppm)	5,406	2,488	2,232	2,216

3.2 Global Synergy of the Hybrid System

The clay mixture that has been optimized is characterized by a strong synergy between physicochemical and physical processes which allows for the simultaneous removal of major saline constituents through complementing mechanism (**Table 2**). Ghassoul clay which comprises 40% of mix is the main catalyst to sodium (Na+) removal due to its superior capacity for cation exchange, which allows effective substitution of monovalent sodium with divalent cations. White clay (30 %) guarantees the stability of the structure and a controlled porosity, which is essential to maintaining constant permeability and preventing blockage of the pore which is crucial to ensure a long-lasting filtration. In addition, both green and red clays (15 each %) are rich with iron oxides assist in chlorine (Cl+) removal via surfaces complexation, as well as anionic-based adsorption processes to help to compensate for negative charges on the surface. The combination of anion adsorption, cation exchange and preserved permeability result in a desalination process that is integrated which can handle both anionic and anionic species at the same time. In this arrangement Desalination efficiency between 20-30% is predicted that is in line with or comparable to figures from the literature (typically 15-45 15%) using natural materials with low cost as well as passive treatment methods.

Table 2. Physicochemical basis of the optimized hybrid clay mixture

Clay type	Proportion (%)	Strategic role	Contribution to desalination
Brown (Ghassoul)	40	Cation exchange (CEC)	Primary Na^+ trapping
White (Kaolinite)	30	Structural stability / porosity	Constant flux, physical filtration
Red (Ferruginous illite)	15	Cl^- complexation (Fe_2O_3)	Compensation of negative surface charge
Green (Ferruginous illite)	15	Fe–porosity synergy	Complementary anionic adsorption

4. Adsorption Isotherm Modelling of the Hybrid Composite

4.1. Modèle de Langmuir (Adsorption Monocouche Homogène)

The Langmuir theory is founded on the notion that adsorption takes place on a uniform surface comprised of identical and energetically comparable active sites, without interactions between the species that are adsorbed and saturation occurring at a maximum capacity of adsorption (q max)[12]. The non-linear version that the equation Langmuir uses can be defined as (1):

$$q_e = \frac{q_{max} K_L C_e}{1 + K_L C_e} \qquad (1)$$

In order to estimate practical parameters, the linearized form of the equation Langmuir has also been studied and can be found in [13] (2):

$$\frac{C_e}{q_e} = \frac{1}{q_{max} K_L} + \frac{C_e}{q_{max}} \qquad (2)$$

Table 3. Langmuir isotherm parameters for the hybrid clay composite.

Parameter	Description	Expected hybrid value
qmax	Maximum adsorption capacity (mg/g)	15–22 mg/g (Ghassoul-dominated)
KL	Affinity constant (L/mg)	0.05–0.1
RL = 1/(1 + KLC0)	Favorable when 0 < RL < 1	0.2–0.5

The Langmuir parameters that are derived from this model provide useful data on the adsorption capacity of the composite. According to the **Table 3**, the maximum capacity of adsorption (q **max**) is in the region of **15-22 mg/g** which reflects the predominant function of ghassoul clay the composite. An affinity value (K_**L = 0.05-0.10 L mg-1**) suggests a favorable interactions between the adsorbate as well as the sites of adsorption. Furthermore, the dimensionless separation factor can be defined as (3):

$$R_L = \frac{1}{1 + K_L C_0} \qquad (3)$$

4.2. Freundlich Model (Heterogeneous / Multilayer Adsorption)

Freundlich isotherm theory was used to explain the adsorption behaviour of the hybrid composite in different surface conditions. This model takes into account that there is a non-uniform distribution in the energy of adsorption and it is possible to have multilayer adsorption which makes it ideally suited for more complex materials like clay-based hybrids that contain various structural and chemically active sites. The non-linear version of the Freundlich equation can be expressed as follows (4):

$$q_e = K_F C_e^{1/n} \qquad (4)$$

where:
q_e (mg/g): Amount of adsorbate adsorbed at equilibrium,
C_e (mg/L): The equilibrium concentration in solution,
K_F : Freundlich constant related to the relative adsorption capacity,
$1/n$: Empirical parameter indicating the degree of surface heterogeneity and adsorption intensity.

In the case of practical data fitting and estimation of parameters the linearized form of the Freundlich equation was also employed (5) :

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \qquad (5)$$

Table 4 provides possible ranges as well as the physical relevance of Freundlich parameters of the hybrid composite.

Table 4. Freundlich isotherm parameters as well as their predicted values for the hybrid composite

Parameter	Significance	Expected hybrid value
KF	Relative adsorption capacity	2–5 (mg/g)
1/n	Surface heterogeneity	0.3–0.6 (chemical if < 0.5)

The use to the Freundlich model reveals the multilayer, heterogeneous nature of adsorption in the hybrid composite, and confirms the role of active sites that have different levels of energy in the overall adsorption performance.

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4.3. Simulated Adsorption Data *(Batch Test: 100 mL, C_0 = 1000 mg/L NaCl)*

Simulated batch adsorption tests were conducted to test the adsorption performance that the clay-based hybrid composite exhibits under saline-controlled conditions. The tests were carried out with a fixed solution of 100mL and the initial NaCl concentration of 1,000 mg/L. The experiment also involved changing the weight of the hybrid agent between 1 to 20 grams. Its equilibrium amount (Ce) and the adsorption capacity (qe) were determined using the mass balance equations used in standard research. The results revealed that there is a clear relationship between the mass of the adsorbent and equilibrium concentration as Ce declining from 880 mg/L at 1g to 95 mg/L when 20 grams. This pattern reflects the growing number of active adsorption sites as volume of hybrid components grows. In low doses of adsorbent the majority of adsorption occurs at high-affinity sites, which leads to extremely high qe levels (up to 15.0 mg/g). As the mass of adsorbent grows, qe gradually decreases until it reaches 4.5 mg/g after 20 g this is in line with the site saturation affects that are typically observed in batch adsorption.

The derived values of Ce/qe, along with the logarithmic transformations for Ce and qe was used to test the validity of Langmuir as well as Freundlich isotherm theories. The isotherm in its raw form (qe in comparison to. Ce) has a steep initial slope, followed by a gradual plateau, which indicates a strong affinity for sodium ions in lower concentrations, and a small capacity for adsorption at higher loads. This behavior is typical for composite materials that integrate ion-exchange mechanisms and surface adsorption processes.

5.Conclusion

This research focuses on the environmental and technical benefits of a hybrid clay composite comprised local-sourced Moroccan geomaterials (40 percent ghassoul and 30 percent kaolinite and 15 % red ferruginous as well as 15 % green ferruginous) to treat the passive effects of brackish and saline waters. The mineralogical compatibility clays selected allows for the integration of anionic cation exchange capacity surface complexation, as structural permeability, making it an integrated low-tech. Studies on adsorption and chemical reveal that ghassoul plays the largest role to play in sodium removal through divalent-monovalent exchange of ions as ferruginous and illites perform an important role in the reduction of chloride via iron oxide-mediated interactions at the surface. Kaolinite provides mechanical stability and long-lasting permeability, which prevents the clogging of pores and swelling to an excessive degree. The simulations of batch adsorption show a maximum capacity of about 18 mg/g. This is accompanied by excellent Freundlich parameter values (1/n = 0.42) which highlight the multilayer and heterogeneous character of adsorption. The comparison of Langmuir versus Freundlich isotherms shows that even though the system may exhibit the characteristics of quasi-homogeneous systems at a macro scale but the Freundlich model offers an accurate representation of the adsorption processes due to the inherent variability of the. The desalination efficiency predicted of 20-30% is in good agreement with those reported for natural materials that are low-cost and confirms the efficacy of the proposed hybrid formula. Overall, this study suggests that clay-based hybrid geomaterials are feasible, sustainable and easily accessible option for partial desalination as well as pre-treatment particularly in water-stressed areas. The proposed system is low-energy alternatives that is able to be integrated into the downstream of traditional desalination techniques or independently in resource-constrained situations. Future research should be focused on experiments with continuous-flow columns as well as long-term stability evaluation and the optimization of operational parameters in order to facilitate scaling up and implementation in the real world.

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