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ARTICLE

Water as the Pore Former in the Synthesis of Hydrophobic PVDF Flat Sheet Membranes for Use in Membrane Distillation

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ABSTRACT

Although PVDF flat sheet membranes have been widely tested in MD, their synthesis and modifications currently require increased use of green and inexpensive materials. In this study, flat sheet PVDF membranes were synthesized using phase inversion and water as the pore former. Remarkably, the water added in the casting solution improved the membrane pore sizes; where the maximum pore size was 0.58 µm. Also, the incorporation of f-SiO2NPs in the membrane matrix considerably enhanced the membrane hydrophobicity. Specifically, the membrane contact angles increased from 96° to 153°. Additionally, other parameters investigated were mechanical strength and liquid entry pressure (LEP). The maximum recorded values were 2.26 MPa and 239 kPa, respectively. The modified membranes (i.e., using water as the pore former and f-SiO2NPs) were the most efficient, showing maximum salt rejection of 99.9% and water flux of 11.6 LMH; thus, indicating their capability to be used as efficient materials for the recovery of high purity water in MD.

1. Introduction

Although water is a crucial component of life, water scarcity and pollution continue to threaten humans, and every living organism and ecosystem on the planet [1,2]. Nearly 1.1 billion people lack access to fresh and clean water [3–5]. These water challenges are commonly observed in rural settlements in developing countries [1,6,7]. Currently, wastewater reuse and desalination serve as suitable alternatives to meet the increasing demand for clean water, and to tackle water shortages. Desalination has been extensively studied using membrane technology [8,9]. Notably, Membrane Distillation (MD) has attracted the interest of many researchers over other membrane technologies due to its efficient removal of salts from water [10–12].

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Membrane distillation is a liquid-mixture separation process through a hydrophobic membrane. The vapour pressure gradient induced by the temperature difference across the two interfaces of the membranes is the main driving force of the process [13–16]. Therefore, the water is transported through the porous membrane in the form of vapour. In addition to process parameters and module designs, MD performance is affected by membrane morphology (i.e., including size and distribution of pores, porosity, and surface roughness), physical and chemical characteristics of the membrane, as well as membrane wettability. Among other challenges, membrane morphology has been the most limiting factor in simultaneously controlling membrane wetting as well as improved fluxes [17–20].

To circumvent the low rates of water recoveries in MD, several studies have focused on the improvement of the membrane pore sizes. Briefly, the electrospinning of nanofibre membranes has been tested as an alternative technique to enhance membrane pore sizes, pore distribution, and porosity [21–24]. However, the resulting membranes were prone to fouling owing to their high surface roughness [10,25–28]. Similarly, highly porous membranes (i.e., pore sizes > 1.5 μm) have shown a negative impact on the Liquid Entry Pressure (LEP) of the membrane. Putting this into a technical perspective, the LEP of membranes must be strictly higher than the MD operating pressures [29,30]. Consequently, nanofibre membranes are likely to encourage the passage of liquid water due to their large pores; thus, compromising the overall performance of the separation process [30,31].

Besides nanofibre membranes, flat sheet and hollow fiber membranes have been extensively explored for their possible application in MD. For instance, Bruggen and co-workers tested commercial PTFE, PVDF, PE, and PP flat sheet membranes in Direct-Contact MD (DCMD)[32]. Water fluxes ranging between 15 – 20 $L \cdot m^{-2} \cdot h^{-1}$ were reported. Several water-soluble pore-forming agents, including polyvinyl pyrrolidone (PVP), polyethylene glycol (PEG), and polyacrylic acid (PAA) have been successfully tested for enhancing the pores of the membrane [33]. Although water is a green component during membrane synthesis, its exploitation in the preparation of MD membranes has been rarely reported. In this study, water was used as the pore former (additive) and non-solvent (coagulant) during the phase-inversion synthesis of hydrophobic PVDF flat sheet membranes for MD. Moreover, octadecyltrimethoxysilane (OTMS)-modified silica nanoparticles (f-SiO_2NPs) were incorporated into the PVDF membranes to enhance their hydrophobicity. This novel combined procedure would highly assist in preventing MD membrane wetting, improving the rate of water recovery, as well as increasing salt rejections.

2. Materials and Methods

2.1 Reagents

PVDF (MW = 534,000 g mol^{-1}), *N,N*-dimetheylacetamide (DMAc) (Puriss p.a., 99.5%) were purchased from Sigma Aldrich (Germany). Deionized water (Direct-Q®, Merck Millipore) were used for solution preparation.

2.2 Synthesis of Membrane Samples

PVDF flat sheet membranes were prepared following a previously reported non-solvent induced phase method [34]. Briefly, PVDF (10 g) was transferred to a conical flask containing DMAc (100 mL). In order to enhance the membrane pore sizes and hydrophobicity, water and OTMS-modified SiO_2NPs were added into the casting solution. The detailed synthesis of OTMS-modified SiO_2NPs is reported elsewhere [35]. The mixtures were stirred for 4 h at 80°C, followed by vacuum degassing. The compositions of the casting solutions are presented in Table 1. The solutions were cast on a glass plate using a casting knife with a gap height of 150 μm, followed by immediate immersion in a 15°C water coagulation bath. The prepared membranes were stored in deionized water for 48 h to complete the phase separation process. The membranes were air-dried prior to characterization and use. Pristine PVDF flat sheet membranes (i.e., neither pore-enhanced nor modified with f-SiO_2NPs) were termed as M1. The pore-enhanced membranes using 2% and 4% water additive were termed as M2 and M3, respectively. Similarly, the pore-enhanced membranes using 2% and 4% water additive and subsequently modified with 1%(w/v) f-SiO_2NPs were termed as M4 and M5, respectively.

Table 1. Composition of the casting solution for the synthesis of PVDF flat sheet membranes

Membrane	PVDF (g)	DMAc (mL)	H_2O (g)	SiO_2NPs (g)
M1	10	100	0	0
M2	10	100	2	0
M3	10	100	4	0
M4	10	100	2	0.1
M5	10	100	4	0.1

2.3 Characterization of Membranes

The surface and cross-sectional morphology of the membranes were investigated using Scanning Electron Microscopy (SEM, JOEL STM-IT300). Likewise, the surface to-

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pography and roughness of the membranes were analyzed using Atomic Force Microscopy (AFM, Witec Alpha 300 A, TS-150). In order to assess the hydrophobic nature of the membranes, water contact angle measurements were conducted using the sessile drop method in a DSA3OE Kruss drop shape analyzer (GnbH). The Liquid Entry Pressure (LEP) was measured using a dead-end cell. The cell was filled with deionized water, and the pressure was gradually increased until the presence of filtrate was recorded. The tensile strengths of the membranes were calculated from the stress-strain graphs, which were obtained using a Small Angle X-ray Scattering system (SAXSpace, Anton Paar GmbH) equipped with a universal extensional fixture. The membrane pore sizes were measured using the dry-to-wet method in a liquid expulsion capillary flow porometer (3G). Finally, the porosity of the membranes was obtained from the polymer density measurements where isopropyl alcohol (i.e., which penetrates inside the pores of the membrane) and de-ionized water (i.e., which does not penetrate through the membrane pores) were used. The following equation was used to calculate the membrane porosity (ε).

$$\varepsilon = \frac{(w_w - w_d)/\rho_i}{(w_w - w_d)/\rho_i + w_d/\rho_p} \qquad (1)$$

Where w_d is the weight of the dry membrane, w_w is the weight of the wet membrane, ρ_i is the density of isopropyl alcohol, and ρ_p is the density of the polymer.

2.4 Membrane performance in Direct Contact Membrane Distillation

The performance of the PVDF flat sheet membranes was studied on a DCMD laboratory-scale set-up using a 35%(w/v) NaCl solution (i.e., typical concentration of dissolved salts in seawater). The temperature of the feed and permeate streams were kept at 60°C and 20°C, respectively. The feed and permeate streams were cycled through the MD cell (i.e., active surface area of 1.25×10^{-2} m^2) at a flow rate of 0.75 $L \cdot min^{-1}$ in a counter flow mode. The feed stream was composed of either deionized water (i.e., resistivity of 18 MΩ.cm) or saline water (i.e. 35%(w/v) NaCl solution), while the initial permeate stream was composed of deionized water. The water flux was calculated using equation 2, by following the change in permeate weight (g) over time. The weight increment of the water passing through the membrane in vapour state was measured using an EMB 3000_1 weighing balance (Kern & Sohn GmbH). To calculate the salt rejection efficiency, the conductivity of the feed and permeate solutions was monitored using a Shimadzu conductivity meter. Based on the initial and final electrical conductivities of the feed and the permeate, equation 4 was used to calculate the salt rejection (*SR*) performance.

$$J_{water} = \frac{\Delta V}{\Delta t \cdot A} \qquad (2)$$

Where ΔV is the volume of the permeate collected at a time interval Δt, and A_m is the membrane surface area. The difference in volume (ΔV) was calculated from the change in mass (Δm) of the water collected, where 0.997 kg/L was used as the density (ρ) of water at room temperature (equation 3).

$$\Delta V = \frac{\Delta m}{\rho} \qquad (3)$$

$$SR = \left(1 - \frac{C_p}{C_f}\right) * 100\% \qquad (4)$$

Where *SR* is the salt rejection, C_f and C_p are the salt concentrations in the feed and permeate streams, respectively.

3. Results and Discussion

3.1 Morphological Analysis of Membranes

The SEM analysis was performed to investigate the surface morphology and pore geometry of the PVDF flat sheet membranes. Figure 1 presents the surface and cross-sectional micrographs of the membranes. All membranes exhibited similar topography with a slight change in pore size when the amount of pore former (water) was increased in the casting solution. The differences in membrane pore sizes were discussed in the forthcoming sections. During the preparation of the casting solution, the maximum amount of water added was 4%(w/w) relative to the solution. Beyond this weight percentage, the casting solution formed a suspension due to the exchange of solvent (DMAc) and non-solvent (water). Remarkably, the porous structure of the membrane was formed when the coagulation processed was slowed down. Therefore, the added water during the preparation of the casting solution reduced the diffusion rate of the non-solvent during phase separation.

A careful analysis of the cross-sectional images demonstrated the formation of finger-like pore structures. Remarkably, the interstitial spaces between the porous internal structures increased with increasing water content (Figure 1 M2-M5). Similar observations were reported by Malik et al. (2018) when water-soluble polymers such as PAA and PEG were used as pore formers during membrane synthesis [33]. Contrarily, the addition of f-SiO_2NPs did not show a significant change in surface and internal morphology. Therefore, the water added as an additive in the casting solution played a significant contribution in the formation of the microvoids within the internal structure of the membrane. According to Gumbi and co-workers, water increases the casting viscosity; thus, bringing the solution viscosity closer to the binodal composition where the suppression of macrovoids is favoured [34]. These changes in membrane morphology as a function of increase in additive content have a direct impact on membrane topology and roughness [36].

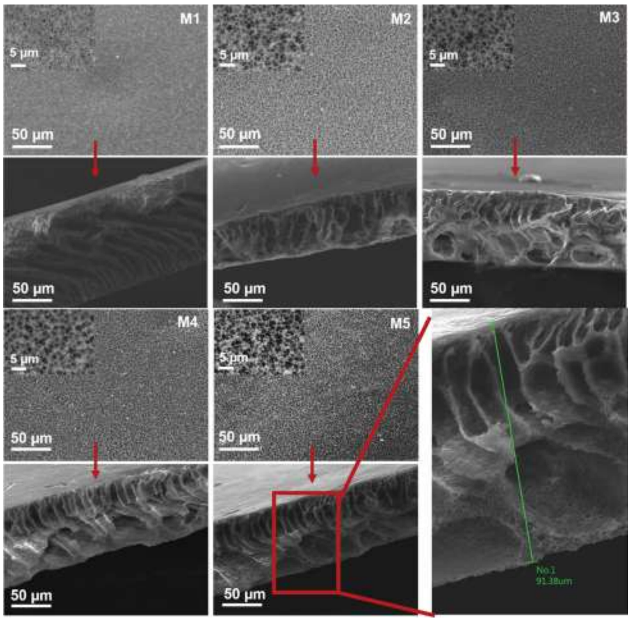

Figure 1. SEM micrographs of PVDF flat sheet membranes

Note: (M1) Pristine PVDF flat sheet membranes, (M2 & M3) pore-enhanced membranes using 2% and 4% water additive, respectively, and (M4 & M5) pore-enhanced membranes using 2% and 4% water additive, respectively, and subsequently modified with 1%(w/v) f-SiO_2NPs.

To investigate the effect of additives (i.e., water and f-SiO_2NPs) on the membrane topology and roughness, an AFM analysis of the membranes was conducted (Figure 2). The analysis was performed on pristine PVDF flat sheet membranes (M1), pore-enhanced membranes using 4% water additive (M3), and pore-enhanced membranes using 4% water additive and subsequently modified with 1%(w/v) f-SiO_2NPs (M5). These membranes selected for AFM characterization are a representative of the different synthesis methods performed in the current study. The surface roughness increased upon addition of water in the casting solution, as evidenced by the root mean square roughness ($Sq_{M1} < Sq_{M2}$). Likewise, the addition of f-SiO_2NPs further enhanced the membrane surface roughness. Thus, the order of membrane surface roughness was M1<M3<M5. Remarkably, the increase in membrane roughness was not only caused by the pore formation induced by water added as an additive in the casting solution but also by the membrane viscosity as stated in the previous section. Additionally, surface roughness was mainly caused by the protrusion of SiO_2NPs leading to the formation of re-entrant structures. In previous studies, it was reported that membranes with high surface roughness encourage air-entrapment [26]. These air-entrapment increases membrane resistance to wetting by low surface tension liquids. An increase in membrane roughness is an indication of increased effective membrane area caused by the nodular shapes with valleys, which is ultimately beneficial for MD applications.

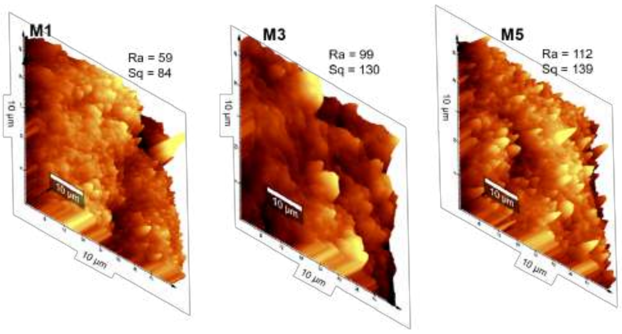

Figure 2. AFM images of PVDF flat sheet membranes

Note: (M1) Pristine PVDF flat sheet membranes without the pore former and SiO_2NPs, (M3) pore-enhanced membranes using 4% water additive, and (M5) pore-enhanced membranes using water 4% water additive and subsequently modified with 1%(w/v) f-SiO_2NPs.

3.2 Contact Angles and Liquid Entry Pressure

The membranes used in MD applications should be strictly porous and not be wetted by the process liquids [10]. Consequently, the LEP should not be lower than the operating pressure of the process. Therefore, the membranes should be hydrophobic. As such, the effect of additives on the membrane wettability (i.e., assessed by the membrane contact angle) and liquid entry pressure were presented in Figure 3. The contact angle (CA) results showed that the pristine PVDF flat sheet membranes (M1) were hydrophobic (CA = 96±2°). The addition of water as the pore former did not impact the membrane contact angles. The contact angles for M1, M2, and M3 were approximately similar ($CA_{M1} = 96°$, $CA_{M2} = 97°$, $CA_{M3} = 94°$). Remarkably, the PVDF membranes changed from hydrophobic to superhydrophobic ($CA \approx 153°$) upon the addition of f-SiO_2NPs. This phenomenon was in agreement with pre-

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viously reported studies [37,38].

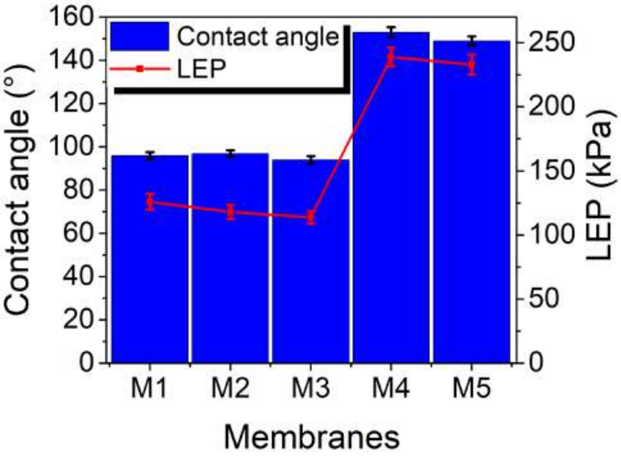

Figure 3. Contact angles and LEP of PVDF flat sheet membranes

Note: (M1) Pristine PVDF flat sheet membranes, (M2 & M3) pore-enhanced membranes using 2% and 4% water additive, respectively, and (M4 & M5) pore-enhanced membranes using 2% and 4% water additive, respectively, and subsequently modified with 1%(w/v) f-SiO_2NPs.

The membrane-water contact angle showed a direct impact on the liquid entry pressure (LEP). Also, the LEP was affected by the membrane pore size as well as the geometry. Notably, the LEP gradually decreased from 126 kPa (M1) to 114 kPa (M3). This change in LEP was ascribed to the increase in membrane pore-size (Figure 4). A drastic change in LEP (i.e. from 126 kPa to 239 kPa) was observed on the superhydrophobic membranes (i.e., compared to the hydrophobic membranes). This change was caused by cavitation induced by the superhydrophobic nature of the membranes and the water polarity. Water becomes energetically unstable when in contact with the superhydrophobic membranes, resulting in the formation of water bubbles and blocking the membrane pores. Consequently, the water requires higher pressures to be driven across the membrane. This phenomenon was also reported by Li and co-workers [39].

3.3 Pore Size and Mechanical Strength of Membranes

As previously indicated, membrane pore sizes have a direct relationship with LEP. Similarly, the membrane pores also exert an impact on the mechanical strength of the membrane. Therefore, to investigate the effect of additives on the size of the pores and the mechanical strength of membranes, the membrane pore size and Young's modulus were recorded and presented in Figure 4. The pore sizes of membranes M1, M2, M3, M4, and M5 were 0.21 µm, 0.39 µm, 0.52 µm, 0.47 µm, and 0.58 µm, respectively. The size of the membrane pores depends on the rate of solution phase separation. Briefly, the addition of the pore former (water) decreased the rate of solvents exchange during phase separation leading to the formation of big pores. Therefore, water was the primary contributing factor for the formation of highly porous membranes.

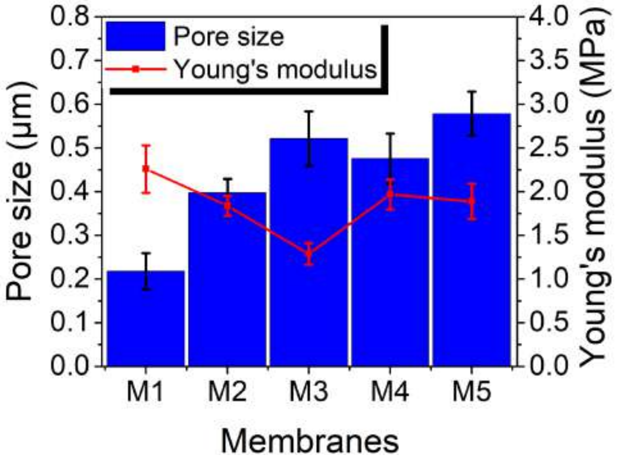

Figure 4. Pore size and Young's modulus of PVDF flat sheet membranes

Note: (M1) Pristine PVDF flat sheet membranes, (M2 & M3) pore-enhanced membranes using 2% and 4% water additive, respectively, and (M4 & M5) pore-enhanced membranes using 2% and 4% water additive, respectively, and subsequently modified with 1%(w/v) f-SiO_2NPs.

While highly porous membranes are a pre-requisite in MD, the increase in membrane pores led to a decrease in mechanical strength, as evidenced by Young's modulus (Figure 4). Specifically, Young's moduli decreased from 2.26 MPa for the pristine membrane (M1) to 1.29 MPa upon the addition of 4% H_2O in the casting solution (M3). This lower mechanical strength was associated with increased voids within the structure, causing a crack propagation during tensile stress and subsequently resulting in a mechanically weaker membrane. However, further addition of f-SiO_2NPs led to a slight increase in mechanical strength. The incorporation of NPs in the membrane decreased the membrane porosity, rendering them stronger. Also, the silica nanoparticles act as crosslinkers in the composite membranes and increase the membrane rigidity, which subsequently improved their mechanical strength. Similar observations have been reported elsewhere [38,39].

3.4 Flux and salt rejection studies

The water flux and salt rejection across all PVDF flat sheet membranes were studied in a DCMD set-up, and the results were presented in Figure 5 and 6, respectively. The measurements were conducted at 10 h. intervals while the feed and permeate temperatures were kept at 60° and 20°, respectively. The initial water fluxes of M1, M2, M3, M4, and M5 were 6.6 LMH, 7.8 LMH, 8.2 LMH, 10.2 LMH, and 11.6 LMH, respectively. An unstable slight increase in water flux on M1, M2, and M3 was associated with the possible wetting within membranes; thus, promoting the passage of water in a liquid state. This possible membrane wetting would be related to low LEP values (114-126 kPa). Thus, the final water flux on M1-M3 ranged between 6.9 LMH – 8.1 LMH. Contrariwise, an unstable slight decrease in water flux was observed on M4 and M5. This flux decay was attributed to the possible temperature polarization and membrane artifacts deviations. The final water flux on M4 and M5 was 9.4 LMH and 9.7 LMH, respectively. Although the superhydrophobic membranes (M4-M5) were affected by the process parameters, resulting in a flux decay as a function of time, they performed well in terms of fluxes compared to the less hydrophobic M1-M3.

The salt rejection studies were performed using 35%(w/v) NaCl. A 99.9% salt rejection was recorded for f-SiO_2NPs-modified PVDF flat sheet membranes. An almost stable salt rejection was observed with a minimal loss (≤0.06%) on M4 and M5. Furthermore, a slight loss in salt rejection (from 99.2% to 97.8%) was recorded on pristine PVDF membrane (M1). Salt rejection losses recorded for M1-M3 were in the range of 1.4-1.5%. The higher decay in salt rejection on M1-M3 compared to M4-M5 would be attributed to membrane wetting. While membrane porosity is a requirement in MD, highly porous membranes with low hydrophobicity promote membrane wetting by the process liquids; thus, compromising the separation efficiency of the membrane. Therefore, a combined strategy for improved membrane porosity and hydrophobicity is imperative.

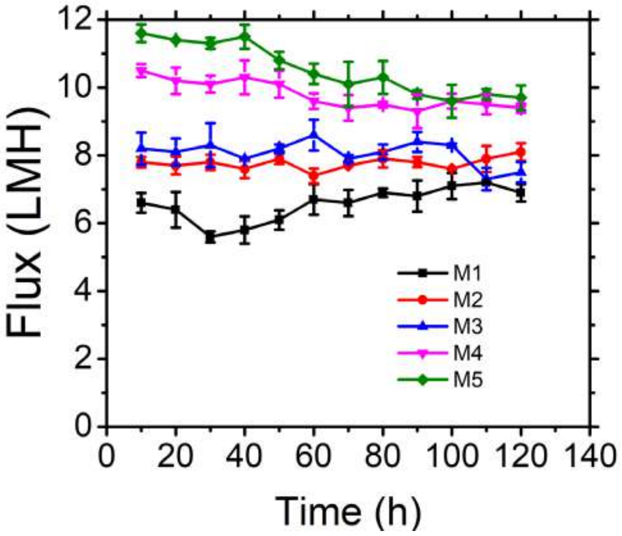

Figure 5. Water flux in DCMD using PVDF flat sheet membranes

Note: (M1) Pristine PVDF flat sheet membranes, (M2 & M3) pore-enhanced membranes using 2% and 4% water additive, respectively, and (M4 & M5) pore-enhanced membranes using 2% and 4% water additive, respectively, and subsequently modified with 1%(w/v) f-SiO_2NPs.

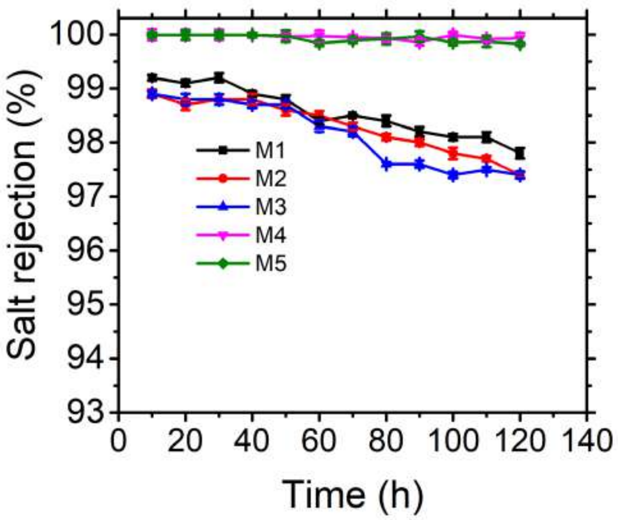

Figure 5. Salt rejection in DCMD using PVDF flat sheet membranes

Note: (M1) Pristine PVDF flat sheet membranes, (M2 & M3) pore-enhanced membranes using 2% and 4% water additive, respectively, and (M4 & M5) pore-enhanced membranes using 2% and 4% water additive, respectively, and subsequently modified with 1%(w/v) f-SiO_2NPs.

3.5 Membrane Performance Comparison Studies

A comparison of MD performance using PVDF flat sheet membranes was illustrated in Table 2. This summary was obtained from the literature and the current study. The synthesized PVDF flat sheet membranes demonstrated performances similar to those in the literature. The highest flux (22.4 LHM) was reported by Fan et al (2013) [40]. However, the salt rejection reported were similar to those obtained from the f-SiO_2NPs reported in the current study.

Table 2. Properties of PVDF membranes and their performances at 60°C feed temperature

Membrane	**Thickness (µm)**	**CA (°)**	**LEP (kPa)**	**Pore size (µm)**	**Porosity (%)**	**Youngs modulus (MPa)**	**Flux (LMH)**	**Rejection (%)**	**Ref.**
M1	101.3	96	126	0.22	76	2.3	6.6	99.2	This study
M2	73.4	97	118	0.39	81	1.8	7.8	98.9	This study
M3	94.2	94	114	0.52	86	1.3	8.2	98.9	This study
M4	90.5	153	239	0.47	78	2.0	10.2	99.9	This study
M5	91.3	141	233	0.58	79	1.9	11.6	99.9	This study
PVDF/SiO_2	-	135	310	-	-	-	2.9	99.9	[41]
PVDF/SiO_2	-	156	275	0.52	69	-	8.3	99.9	[42]

PVDF/ PTFE	81	145	120	0.43	80	-	22.4	99.9	[40]
PVDF	62	-	300	0.34	79	-	5.2	-	[43]
PVDF	104	87	-	0.48	54	-	12	-	[44]
PVDF/ LiCl	-	107	290	-	81	-	9.7	99	[45]

4. Conclusion

In this study, highly porous and superhydrophobic PVDF flat sheet membranes were synthesized and tested for water desalination in MD. The addition of water in the casting solution resulted in improved membrane pores. To achieve superhydrophobicity, f-SiO_2NPs were incorporated in the flat sheet nanocomposite membranes. Remarkably, the NPs did not affect the membrane pore sizes, clearly demonstrating the combined achievement of membrane pores and hydrophobicity for improved application in MD. The recorded membranes pore sizes, mechanical strength, LEP, contact angle were in the following ranges; 0.21-0.58 μm, 1.29-2.26 MPa, 114-239 kPa and 96-153°, respectively. These membranes were efficient in salt rejection. However, the f-SiO_2NPs-modified membranes showed the highest efficiencies. Furthermore, the modified PVDF membranes were characterized by high water fluxes, indicating a promising use for high rate water recoveries in MD.

Declaration

The authors declare no conflict of interest.

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ARTICLE

Groundwater Drainage By Means Of Electrochemistry For Soil Improvement

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ABSTRACT

In the last decade, the construction of communication routes has intensified globally. As a result, many case studies have emerged related to the presence of saturated clayey soils in the foundation ground. In order to speed up the execution of highways and railways in a safely manner, the designers use different methods for improving soft clays in terms of compressibility. The present paper aims to evaluate the efficiency of an electrochemical method used for the dewatering of a soft montmorillonite clay subgrade. The effects on the consolidation process are analyzed for the final conclusions. The advantages of the method are briefly discussed, and some potential areas for scientific research are proposed.

1. Introduction

In applying the methods of soil improvement, it is necessary to understand the soil behavior and the natural phenomena that may affect it. A distinction must be made between the concept of soil consolidation as a natural phenomenon of compression under a certain load in time and the methods for consolidating the foundation ground through artificial improvements.

The electro-osmotic consolidation is a soil improvement method during which the pore water in the soil is drawn from an anode to a cathode under an induced electric field, thus creating a negative pore pressure. This phenomenon leads to the soil consolidation and the increase of the soil's strength parameters. This increase has proven to be permanent in clayey soils.

The electro-osmosis was first used by Casagrande [1] in geotechnical engineering and since then, it has been used in various applications, including stabilization of slopes, dams and embankments, soil improvement, dewatering of sludge and tailings, remediation works and groundwater displacement.

The combination of electro-osmosis and other techniques (preloading, chemical treatment, lime columns, vacuum) was also studied and numerous publications emerged over time.

The electro-osmosis can also be used as a tool in predicting the soil behavior during consolidation.

A laboratory experimental stand was developed for observing and evaluating the electro-osmotic consolidation process in a soft montmorillonite clay sample, as well as the variation of the mechanical properties of the clay be-

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fore and after the experiment.

2. Methodology and materials

2.1 Methodology

The total stress in any saturated soil, σ, is comprised of two parts, namely the effective stress, σ', and the pore water pressure, u, as follows:

$$\sigma = \sigma' + u\left[kPa\right] \tag{1}$$

The pore water pressure, u, is obtained by multiplying the unit weight of the water, γ_w, with the piezometric head, h_w:

$$u = \gamma_w \cdot h_w\left[kPa\right] \tag{2}$$

When an electrical current is applied across a saturated soil mass, the cations migrate toward the cathode and the anions toward the anode. The cations containing negatively charged particles from a clayey soil are in greater number, thus causing the water to migrate toward the cathode. This phenomenon is called electro-osmosis and its efficiency depends on a series of factors, such as the conductivity of soil, the voltage gradient, and the water properties.

During the electro-osmosis, a negative pore pressure is induced, leading to an increase of the effective stress, thus resulting the consolidation of the soil.

$$\sigma' = \sigma - \left(-u\right) = \sigma + u\left[kPa\right] \tag{3}$$

When the pore water is drawn from the anode towards the cathode, a hydraulic gradient is developed between the electrodes and there is free drainage at the cathode [2].

The relationship below is frequently used in geotechnical engineering for evaluating the electro-osmotic flow, being applicable when there is free access to water at both the anode and the cathode:

$$q = k_e \cdot i_e \cdot A\left[\frac{m^3}{s}\right] \tag{4}$$

, where:

k_e = coefficient of electro-osmotic permeability [m^2/sV or m/s per V/m]

i_e = electrical gradient [V/m]

A = cross-sectional area perpendicular to water flow [m^2]

This method acts on the weakly bound (film) water within the adsorption complex and especially on the free water.

Because cations of different valence (eg. Na^+, Ca^{2+}) may exist in the adsorption complex, the efficiency of the method also depends on the chemicals dissolved in the pore water of the soil. For example, marine clays (eg. Singaporean clays) are different from clays formed in freshwater.

2.2 Materials

The material used to carry out the experiment was a saturated montmorillonite clay, with the basic properties listed in Table 1.

Table 1. Basic properties of the clay used in the experiment

Parameter	Initial water content, w (%)	Degree of saturation, S (%)	Liquid limit, w_L (%)	Plastic limit, w_P (%)	Specific gravity, ρ_s (g/cm^3)
Value	46.94	100	72.30	23.83	2.70

Before and after the experiment, the material was subjected to consolidation tests in the edometer to observe the effect of the electro-osmosis phenomenon on its mechanical properties (Figure 1).

Figure 1. Mounting of the sample in the edometer apparatus

After edometric consolidation on different loading steps (200, 300 and 500 kPa), the values presented in Table 2 were obtained.

The clay was placed in a plastic container of 27x18x12 cm in size.

The electrodes used (the anode and the cathode) consisted of two carbon iron bars (OB 37), which were inserted into the midline of the soil sample, at a distance of 17 cm, resulting a voltage gradient of 1.03 V/cm.

The power source consisted of a car rectifier, with a power supply at 220 V/50 Hz, the resulting voltage being

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12 V/DC. A multimeter was used to monitor the electrical current.

The total testing time was 72 hours.

The experimental stand is shown in Figure 2.

Table 2. Consolidation parameters of the clay

Stress, (kPa)	Deformation, e (%)	Compression modulus, M (kPa)	Coefficient of volume compressibility, m_v (kPa^{-1})	Coefficient of consolidation, c_v (cm^2/s)	Coefficient of permeability, k (cm/s)
100	1.45	2688	0.000372	4.978E-06	1.952E-06
200	5.17				
200	5.17	3125	0.00032	3.795E-06	1.325E-06
300	8.37				
300	8.37	3756	0.0002663	3.395E-06	1.047E-06
500	13.69				

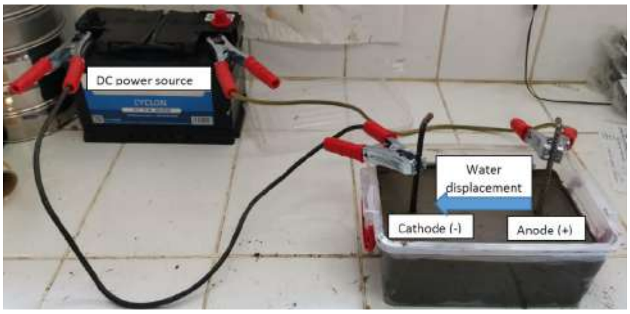


Figure 2. Experimental stand

3. Results and Discussion

After about 2 hours from the beginning of the experiment, the formation of a water film around the cathode was observed (Figure 3a), thus attesting the occurrence of the electro-osmosis phenomenon.

During the electro-osmotic consolidation, the water moved from the anode to the cathode inside the soil sample, causing the formation of some cracks near the anode (Figure 3b).

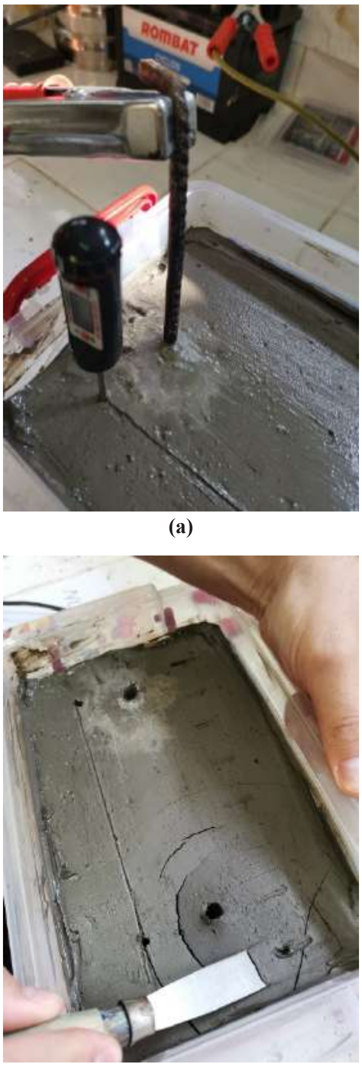
During the experiment, the following aspects were observed:

(1) the temperature measured near the cathode with the help of a thermometer recorded values with approx. 2 degrees higher than the one next to the anode throughout the experiment;

(2) the anode underwent oxidation during electro-osmosis, losing approx. 0.5% of the initial mass (Figure 4);

(3) the humidity of the soil sample near the cathode increased by approx. 14% compared to that determined near the anode;

(4) approx. 200 ml of water were discharged from the clay sample.



(a)

(b)

Figure 3. Experimental stand after the experiment: (a) formation of the water film around the cathode; (b) cracks appearance around the anode

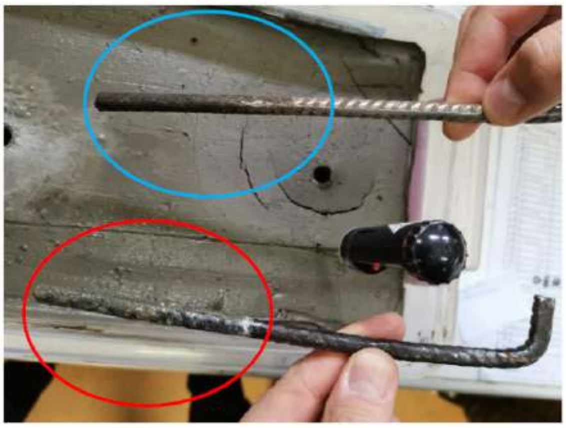


Figure 4. Oxidation effect of the anode (red circle) compared to the cathode (blue circle)

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Taking into account the scale of the model, we consider that the effect of applying the electro-osmosis method on the soft clay is more than satisfactory.

In order to directly point to this conclusion, mechanical compressibility tests were performed on the clay sample improved by electro-osmosis.

After edometric consolidation on different loading steps, the values presented in Table 3 were obtained.

Table 3. Consolidation parameters of the clay after electro-osmosis

Stress, (kPa)	Deformation, e (%)	Compression modulus, M (kPa)	Coefficient of volume compressibility, m_v (kPa^{-1})	Coefficient of consolidation, c_v (cm^2/s)	Coefficient of permeability, k (cm/s)
100	3.00	5952	0.000168	6.624E-06	1.246E-06
200	4.68				
200	4.68	5602	0.0001785	4.217E-06	8.574E-07
300	6.47				
300	6.47	5831	0.0001715	3.488E-06	7.084E-07
500	9.90				

The diagrams in Graphs 1-3 show the differences recorded before and after the application of electro-osmosis on the clay sample. From these graphs and from the values presented in Table 3, some very interesting engineering conclusions are drawn, namely:

(1) the compression-consolidation curves highlight the effect of "speeding up" the consolidation process and the increase of the specific deformations' values for the samples along with the variation of the consolidation pressure; if we consider the degree of vertical consolidation u_v=50% under the conditions of the double drainage specific to the edometric test it is observed that:

(A) for σ=200 kPa, $\Delta\varepsilon_{50}\approx1\%$;

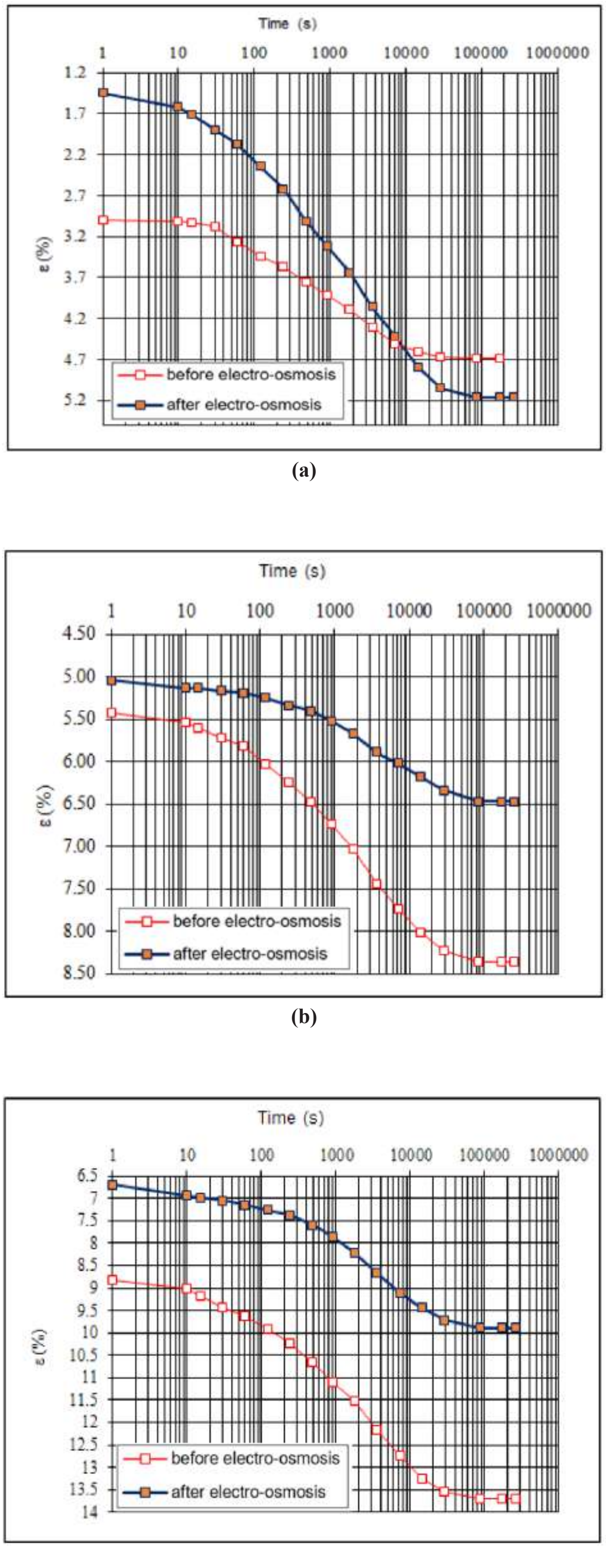
(B) for σ=300 kPa, $\Delta\varepsilon_{50}\approx1,5\%$;

(C) for σ=500 kPa, $\Delta\varepsilon_{50}\approx3\%$.

(2) the effects of the pressure, σ, on the consolidation coefficient's variation, c_v, is approx. 2.5 times higher for the clay improved by electro-osmosis in the range of (200÷300) kPa;

(3) the permeability coefficient, k, decreases to a range of values between ~ $0.75x10^{-6}$ cm/s and ~ $0.3x10^{-6}$ cm/s for loads within the range of (200÷500) kPa;

(4) it turns out that by applying the electro-osmosis process in soft clays deposits (very plastic and active in relation to water), the obvious improvement of the clay's mechanical properties (the compressibility decrease under increasing pressures) takes place.



(a)

(b)

(c)

Graph 1. Edometric consolidation curve under different loading stages: (a) σ=200 kPa; (b) σ=300 kPa; (c) σ=500 kPa

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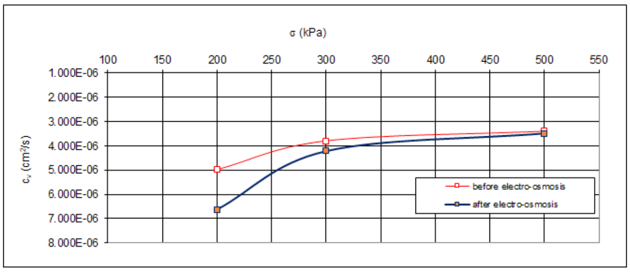
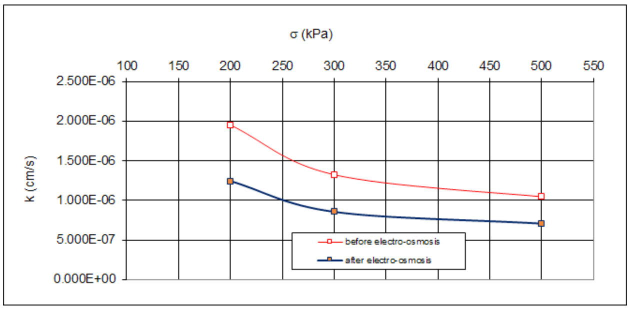


Graph 2. The variation of the consolidation coefficient with pressure



Graph 3. The variation of the permeability coefficient with pressure

4. Conclusion

The obtained results showed the electro-osmosis' influence on the soft clay's properties. The electrical current allowed the fast drainage of water from the clay sample.

An obvious increase of the clay's coefficient of consolidation was recorded, as well as a decrease of the permeability coefficient. All the measured values were within those provided by literature, confirming the efficiency of the electro-osmotic process.

Nevertheless, the results also showed the influence of electro-osmosis on the flow of water, a phenomenon that can successfully be used for remediation of contaminated soils and other similar applications.

The conductivity of the material plays an important role in controlling the efficiency of the electro-osmosis method. The increase in conductivity leads to a decrease in resistivity, thus improving the water flow through the sample.

Studies have shown that saline water added in the soil sample instead of tap water led to an increase in permeability by a factor of 1.2, without any electrical current connection [3].

Therefore, an interesting future research would regard the interaction between electro-osmosis and groundwater with a high salt content. This method could be very useful for coastline consolidation applications.

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ARTICLE

Rain Water Characterization at Urban and Rural (North B) Unguja

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Rainwater

ABSTRACT

Aim of the study was to assess the physicochemical characteristics of rainwater at urban and rural (north B) Unguja. Twenty four (24) rainwater samples were analyzed for pH, electrical conductivity (EC) and total dissolved solids (TDS) and cations: calcium (Ca^{2+}), copper (Cu^{2+}), iron (Fe^{2+}), chromium (Cr VI) , and anions: fluoride (F^-), nitrite and sulfate (SO_4^{2-}). The investigated physicochemical parameters in the rainwater samples were within the permissible limits of the World Health Organization (WHO) drinking water quality guidelines. The ranges of concentration for the physical parameters analyzed were; pH: (5.8 – 7.7), EC: (5.08 – 35.6 µS/cm), TDS: (1.68 – 20.16 mg/L, Ca: (0.163 – 24.331 mg/L), Cr: (VI) : (BDL – 0.029 mg/L), Cu: (BDL - 0.085 mg/L); Fe: (BDL – 0.068), anions were SO_4^{2-} : (BDL – 5.00 mg/L) and F- : (BDL – 0.5 mg/L). Nevertheless, there was no significant difference for the measured parameters between urban and rural areas. This is a good indication that the air quality in Zanzibar is remarkably not polluted with respect to the parameters studied.

1. Introduction

Water is among the primary needs of life, playing important roles including human consumption, industrial processing, irrigation and domestic waste water management [1]. The human existence and physiology depend much on availability of good quality water [2]. Safe drinking water is more important for human life, however much of the world's population does not have access to safe drinking water [3].

The growing of human population and other living organisms threaten the water resources due to demanding the quality water for domestic and economic activities purposes [4]. Decision-makers faces significant challenges on the issues of supplying adequate water to the rapidly growing population especially in most urban area, where by the societal needs and equity on water access is required.. However the water quality changes during processes like harvesting, storage and house hold use [5].

Rainwater resources and others like river, sea and ground water are on a great serious threat from over uses or pollution due to anthropogenic activities or sources like combustion of fuels for energy generation, heating, transport and industrial needs, previously polluted and waste disposal sites, wind blow soils from arid and agricultural regions [6,7].

Acid rain is referred as any rain fall containing excessive amount of dissolved acid gases like SO_2, NO_2 and CO_2 and particulate matter in the atmosphere, rain water becomes acidic if the pH is less than 5.5 when it falls to the surface [8].

Rainwater is a free source that can be obtained naturally and considered to be the purest form of water which formed through natural distillation. This water contains dissolved

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gases such as nitrogen dioxide, ammonia, sulphur dioxide, aerosol or particulate matter from atmosphere [9,10,11]. Rainwater acts as a collector which is used to detect and control the concentration of gaseous and particulate matter that includes the trace metal in the atmosphere [12].

Remarkably, water reserve has been found to be in short supply globally; the water shortage has sent man on the lookout for new alternative water resources of rainwater harvesting that may take the role of intake of number of nutritional and toxic trace elements in man. [13]. Therefore, this study aims at determining the concentration of cations (Ca^{2+}, Cu^{2+}, Fe^{2+}, Zn^{2+}, Cr (VI) and anions (F^-, NO_2^-, SO_4^{2-}) at selected areas of Urban and rural (north B) in Zanzibar Island. Other parameters such as EC, pH and TDS were also determined. Moreover, comparison of the level of these ions was made between Urban and Rural areas. Nevertheless, the measured parameters in rainwater were compared to WHO guidelines to ascertain its suitability for human consumptions.

2. Materials and Method

2.1 Study Area

The study was conducted at urban west region and North B District of Unguja Island in Zanzibar. Rainwater samples were collected at fourteen (14) locations in urban west region and ten (10) locations in rural North B district. The geographical positioning system GPS was used to locate the sampling station (Appendix 1 and Figure 1).

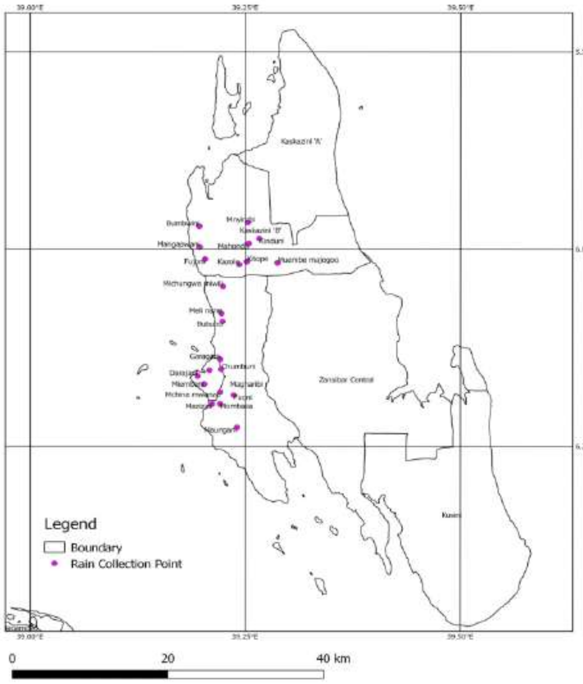

Figure 1. Rain water collection points

2.2 Samples Collection and Storage

On November and December 2016, twenty four rainwater samples were collected from different sites of urban and rural north B areas of Unguja Zanzibar. Immediately after collection, the pH, TDS and conductivity of rainwater samples were measured using HI19812 pH/EC/TDS METER. The handling, storage and preparation of the samples were done in accordance with the standard EPA method 2007.

2.3 Methodology

The analytical and laboratory works by using appropriate standard method were carried out in the Chief Government Chemist Laboratory Agency (CGCLA) Zanzibar and University of Dar- es - salam (UDSM) Tanzania. Table 1 depicts summary of the equipments/ instrument used and parameters analyzed.

Table 1. Parameters Analyzed and the Instrument Used

Parameters	Equipments/Instrument	Test methods
Electrical conductivity (EC), Total dissolved solid (TDS) and pH	HI 19812 pH/EC/TDS METER	Insitu measurement
Sulfate (SO_4^{2-})	UV-1800PC Spectrophotometer	Turbid metric method
Chromium (Cr (VI), copper Cu^{2+} and Fluoride (F^-)	HI 83099 COD and Multiparameter photometer	ASTM D1687-92, DiphenylcarbohydrazideEPA and SPADNS method
Calcium (Ca^{2+}), Iron (Fe^{2+})	Atomic Absorption Spectrophotometry (AAS)	EPA method 2007

2.4 Statistical Analyses

SPSS version 20 was used for the data analysis. The descriptive statistics and Pearson's correlation significant used to examine correlation between selected parameters in water samples is summarized in Table 2.

Table 2. Summary of the Descriptive Statistics of the Analyzed Parameters in Rainwater

Parameters		Mean	Minimum	Maximum	P value	WHO GUIDLINES
pH	1	6.864	6.4	7.7	0.001	6.5 – 8.0
	2	6.290	5.8	7.1		
EC	1	13.848	5.10	33.00	0.902	1500
	2	14.288	5.08	35.60		
TDS	1	7.545	2.415	16.11	0.863	500
	2	7.912	1.68	20.16		
F-	1	0.1057	BDL	0.5	0.56	1.5
	2	0.750	BDL	0.25		

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SO_4^{2-}	1	1.507	BDL	4.5	0.926	400
	2	1.570	BDL	5.0		
Fe^{2+}	1	0.0127	BDL	0.0680	0.132	0.3
	2	0.0012	BDL	0.0037		
Cu^{2+}	1	0.035	BDL	0.18	0.217	2.0
	2	0.021	BDL	0.052		
Cr VI	1	0.00057	BDL	0.003	0.034	0.05
	2	0.0097	BDL	0.040		
Ca^{2+}	1	3.529	0.49	24.331	0.854	75
	2	3.136	0.163	8.410		

Note: 1 - Represent urban areas 2 - Represent rural areas.

3. Results and Discussion

The comparison of the levels of the analyzed parameters between the present study with other studies is highlighted in Table 3.

Table 3. Parameters comparison between the present study with other studies

Parameters	Present study (Parameters Range)	Other Studies (Parameters Range)
PH	5.8 – 7.7	6.22 – 6.92 [14], 6.62 [15].
EC	5.08 – 35.6	14.39 – 43.18 [14], 13.45 – 19.04 [15].
TDS	1.68 – 20.16	7.0 and 25.3 [16], 0.05 – 51 [17]
F^-	BDL – 0.5	0.54 -0.86 [15], 0.07 - 0.48 [18]
SO_4^{2-}	BDL – 5.0	0.057 – 0.396 [19], 2.00-19.00 [20], 3.22 – 7.83 [21].
Fe^{2+}	BDL – 0.068	0.15 – 0.79 [15], 0.6 – 2.2 [14], 17 - 82 [22]
Cu^{2+}	BDL – 0.085	BDL – 0.0009 [14], 0.0028 - 0.0139 [22], 0.0399 - 0.1506 [23].
Cr (VI)	BDL – 0.029	0.00047 – 0.0017 [24]
Ca^{2+}	0.168 – 24.331	0.6 – 0.8003 [14].

Note: All parameters expressed in mg/L except EC (μS/cm), pH (no unit).

3.1 pH

In this study, It was observed that the pH values in rainwater samples varied from 5.8 to 7.7 (Table 3, Appendix 2 and Figure 2) with a mean pH of 6.86 for urban zone, and pH 6.29 for rural areas. There was significant difference for pH (0.001, $P<0.05$) between the urban and rural sites. The lowest pH was recorded at Michungwa miwili, and highest at Darajani. The pH range was slightly higher as compared with similar study in Unguja, Zanzibar (Table 3). Nevertheless, the pH of the present study corresponds to other study whose mean pH value was 6.62 (Table 3). The lower pH value obtained from michungwa miwili (north B) Unguja possibly due to spatial and temporal variation in the atmospheric chemical composition. Moreover, anthropogenic sources, such as agriculture could be a sound reason.

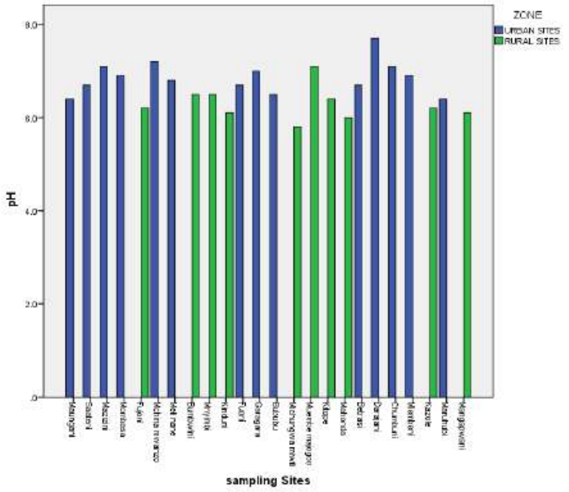

Figure 2. pH of rainwater samples

3.2 Electrical Conductivity (EC)

The lowest conductivity value (5.08 μS/cm) recorded was at Mchina mwanzo, and highest (35.6 μS/cm) at Bumbwini (Figure 3), this could be due to accumulation of various particles of dust, soil, and airborne aerosols that dissolve prior to deposition [25]. Notably, there was no significant difference (0.902, $P<0.05$) for EC between the urban and rural sites. The range of EC is lower as compared with similar study conducted in urban west region of Zanzibar Island (Table 3). Low level of electrical conductivity (EC) in rainwater is an indication that the atmosphere has not been highly scavenged by ions containing aerosols. This finding is consistent with rainwater conductivity level from Tamale metropolis (Table 3).

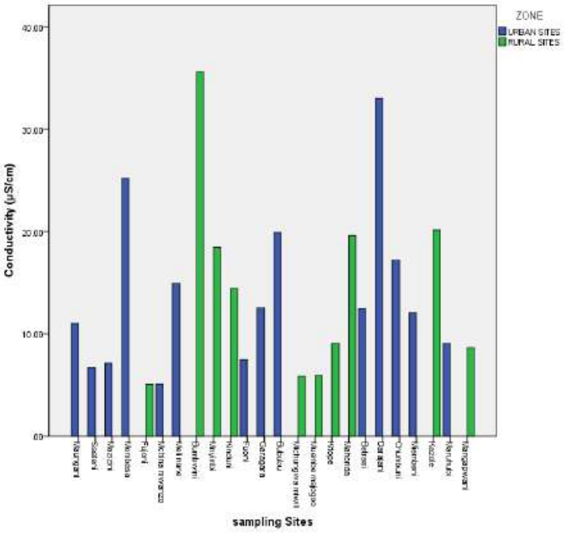

Figure 3. Electrical Conductivity of rainwater samples

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3.3 Total Dissolving Solids (TDS)

TDS values varied from 1.68 to 20.16 mg/L (Table 2, Appendix 2 and Figure 4) with mean of 7.545 and 7.912 ppm at urban sites and at rural sites respectively. There was no significant difference (0.863, $P<0.05$) of TDS between the urban and rural sites. The lowest value obtained for TDS was at Muembe majogoo and highest found at Kazole, this high value reported could be due to the suspended of particulate matter in atmosphere. The finding is consistent with the studies reported at Kotei Ghana and Tamale municipality (Table 3), but all values were within the WHO safe limits (500 mg/L).

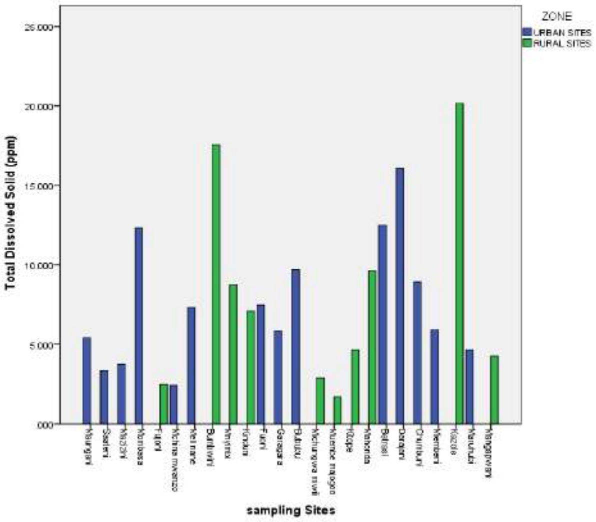

Figure 4. TDS in rainwater samples

3.4 Fluoride

The concentration of fluoride was found to range from BDL to 0.5 mg/L with a mean of 0.1057 mg/L at urban sites and 0.750 mg/L at rural sites (Table 2, Appendix 2 and Figure 5). All samples concentrations were within WHO limits (1.5 mg/L) and basically there was no significant difference (0.56, $P<0.05$) of fluoride between the urban and rural sites. The lowest concentrations of fluoride were detected at Mangapwani, Maruhubi and Fujoni while the highest was detected at Mazizini. This may be due to the occurrence of fluorine in atmospheric air. Also burning of plastic use fluoride-containing substances could pollute the environment with fluoride gas, fumes. or smoke contribute to the environmental fluoride pollution [18], which inturns can contaminate rain water. The fluoride of the present study corresponds to other studies conducted at Ayanfuri, Ghana and Bangladesh (Table 3).

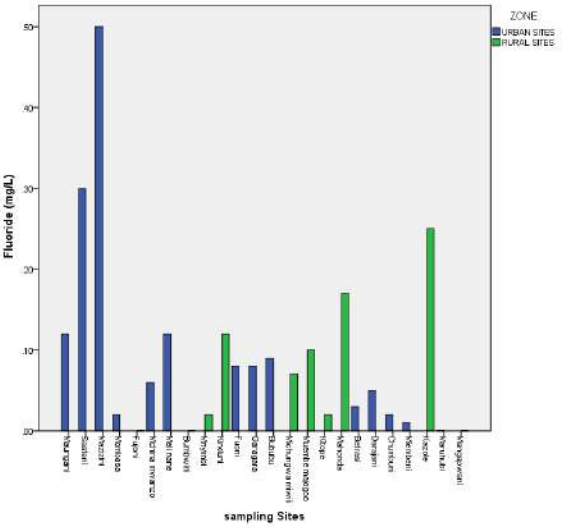

Figure 5. Fluoride level in rainwater samples

3.5 Sulfate

As depicted from Figure 6, Appendix 2 and table 2, the SO_4^{2-} concentration varied from BDL (At Mangapwani, maruhubi Kazole, Miembeni, Fuoni and Chumbuni) to 5 mg/L (at Bumbwini) with a mean of 1.507 mg/L at urban sites and 1.57 mg/L at rural sites. The study didn't show any significant difference (0.920, $P<0.05$) of sulfate concentration between urban and rural sites selected. This could be anthropogenically attributed via burning of sugarcane, soil dust or industrial emission at Mahonda sugar factory. The result collected was below the WHO recommended level of 400mg/L. This study to some extent corresponds to other similar studies. Conducted at different areas (Table 3).

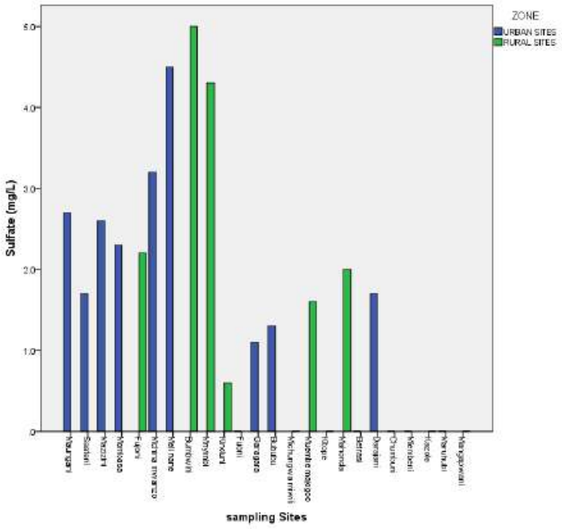

Figure 6. Sulfate level in rainwater samples

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3.6 Iron

Iron concentrations ranged from BDL to 0.068 mg/L with a mean of 0.0127 mg/L at urban sites and 0.0012 mg/L at rural sites. All recorded concentrations were below the WHO limit of 0.3 mg/L. The concentrations of iron were below detection limit for over sixty percent of sampling sites (Table 2, Appendix 2 and Figure 7). Highest concentration of iron was measured at Mazizini and Maungani this could be contributed from agricultural practices. There was no significant difference (0.132 $P<0.05$) of Iron between the urban and rural sites. The iron concentration range of this study was lower as compared with similar studies conducted on other areas (Table 3).

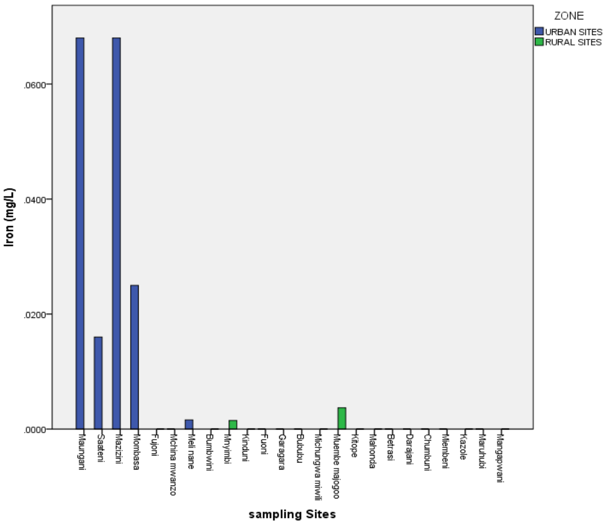

Figure 7. Iron level in rainwater samples preferred

3.7 Copper

The concentration of copper ranges in rainwater samples from BDL – 0.18 mg/L and BDL – 0.052 mg/L at urban and rural areas respectively (Table 2, Appendix 2 and Figure 8). They were below the WHO guidelines (2.0 mg/L). The concentration of copper between the urban and rural sites didn't show any significant difference (0.217, $P<0.05$), where the highest value of 0.18 mg/L was recorded at Saateni (sample number 2), and the lowest concentration recorded bellow the detection limit at Bumbwini, Kazole, and Mangapwani (samples number 8, 22 and 24 respectively). Basically, the value of copper reported in this study is comparable with other studies (Table 3).

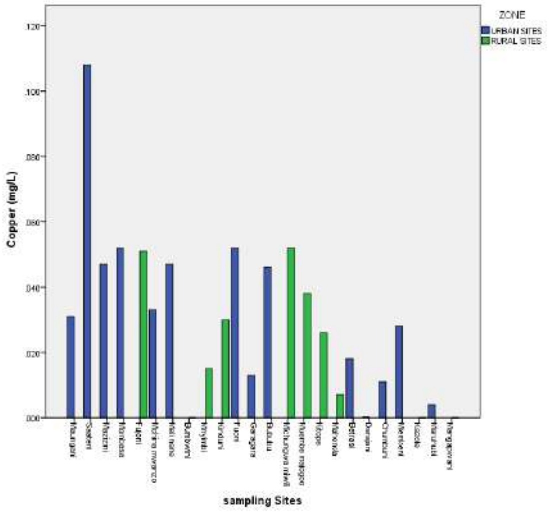

Figure 8. Copper level in rain water samples

3.8 Chromium

The Cr (VI) in some rainwater samples was below the detection limit, and maximum concentration was 0.029 mg/L (Table 2, Appendix 2 Figure 9), which was lower than the WHO permissible limit (0.05 mg/L). A comparison of results of chromium in rainwater samples affirmed that there was a significant difference (0.034, $P<0.05$) between the urban and rural sites. The study conducted at peri-urban region of Kleinmond and Western Cape (Table 3) show good correspondence with the chromium data of this study. The high level of chromium at rural sites could be attributed from industrial activities at mahonda sugar factory.

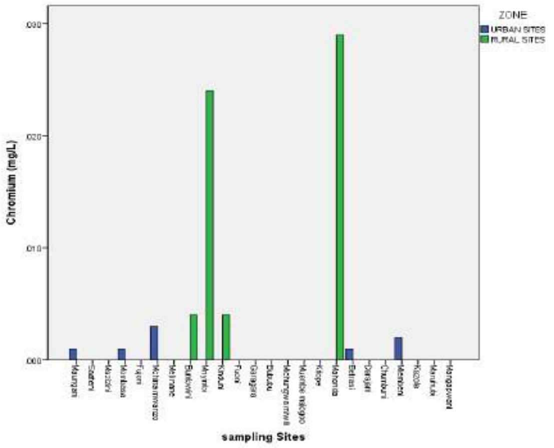

Figure 9. Chromium level in rain water samples

3.9 Calcium

The mean concentration of calcium in rainwater samples were 3.529 mg/L and 3.136 mg/L at urban and rural areas

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respectively (Table 2, Appendix 2 and Figure10. There was no significant difference (0.854, $P<0.05$) of calcium between the urban and rural sites. The lowest value of calcium (0.163 mg/L) was found in Fujoni. and the highest concentration of calcium (24.331 mg/L) was obtained in sample collected from Maruhubi, this could be attributed with the building materials of five star hotel, which is located nearby sample collection point. All data of calcium observed were below the WHO limit (75 mg/L), (Table 3).

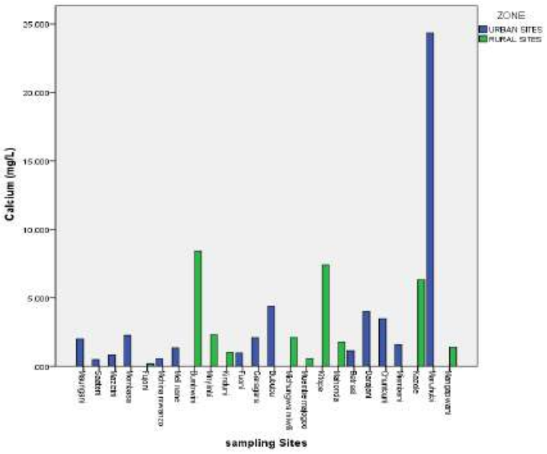

Figure 10. Calcium level in rainwater samples

4. Conclusion

In this study there was no significance difference of the data for almost all parameters analyzed between urban and rural sampled sites except for the pH which observed to be low at rural sites compared to urban site; this could be contributed with sugar factory operations including sugarcane burning. The investigated physico-chemical parameters (pH, EC and TDS) and (Ca^{2+}, Cu^{2+}, Fe^{2+}, Cr VI), F^- and SO_4^{2-}) in the rainwater samples from urban and rural (north B) in Zanzibar Island were found below the guidelines for drinking waters given by the World Health Organization (WHO), making the rainwater suitable for drinking and recreational purposes. The best quality of rainwater selected points can be easy used as primary information for the assessment of air pollution.

Appendix

Appendix 1. Sampling Stations with GPS

S.No	Sampling Sites	Station ID	Coordinate S	Coordinate E
1.	Maungani	S01	6˚13.582’	39˚14.404’
2.	Saateni	S02	6˚09.232’	39˚12.490’
3.	Mazizini	S03	6˚11.789’	39˚12.676’
4.	Mombasa	S04	6˚11.774’	39˚13.227’
5.	Fujoni	S05	6˚00.770’	39˚12.183’
6.	Mchina mwanzo	S06	6˚10.893’	39˚13.234’
7.	Meli nane	S07	6˚04.911’	39˚13.336’
8.	Bumbwini	S08	5˚58.273’	39˚11.791’
9.	Mnyimbi	S09	5˚57.987'	39˚15.165'
10.	Kinduni	S10	5˚59.210'	39˚15.964'
11.	Fuoni	S11	6˚11.119’	39˚14.181’
12.	Garagara	S12	6˚08.391’	39˚13.243’
13.	Bububu	S13	6˚05.520’	39˚13.409’
14.	Michungwa miwili	S14	6˚02.849’	39˚13.433’
15.	Muembe majogoo	S15	6˚01.042'	39˚17.234'
16.	Kitope	S16	6˚00.956'	39˚15.101'
17.	Mahonda	S17	5˚59.587'	39˚15.230'
18.	Betrasi	S18	6˚07. 043’	39˚12.700’
19.	Darajani	S19	6˚09.648’	39˚11.674’
20.	Chumbuni	S20	6˚09.155’	39˚13.316’
21.	Miembeni	S21	6˚10.294’	39˚12.127’
22.	Kazole	S22	6˚01.182’	39˚14.568’
23.	Maruhubi	S23	5˚57.987'	39˚15.165'
24.	Mangapwani	S24	5˚59.852’	39˚11.818’

Appendix 2 (A). Levels of the analyzed parameter in rainwater samples (cations, anions, TDS, in mg/L, and EC in µS/cm)

sampling Sites	SO_4^{2-}	Cu^{2+}	Cr (VI)	Fe^{2+}	ZONE
1	2.7	0.031	0.001	0.068	1
2	1.7	0.108	BDL	0.016	1
3	2.6	0.047	BDL	0.068	1
4	2.3	0.052	0.001	0.025	1
5	2.2	0.051	BDL	BDL	2
6	3.2	0.033	0.003	BDL	1
7	4.5	0.047	BDL	0.0016	1
8	5	BDL	0.004	BDL	2
9	4.3	0.015	0.024	0.0015	2
10	0.6	0.03	0.004	BDL	2
11	BDL	0.052	BDL	BDL	1
12	1.1	0.013	BDL	BDL	1
13	1.3	0.046	BDL	BDL	1
14	BDL	0.052	BDL	BDL	2
15	1.6	0.038	BDL	0.0037	2
16	BDL	0.026	BDL	BDL	2
17	2	0.007	0.029	BDL	2
18	BDL	0.018	0.001	BDL	1
19	1.7	0.0003	BDL	BDL	1
20	BDL	0.011	BDL	BDL	1
21	BDL	0.028	0.002	BDL	1

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22	BDL	BDL	BDL	BDL	2
23	BDL	0.004	BDL	BDL	1
24	BDL	BDL	BDL	BDL	2

Appendix 2 (B). Levels of the analyzed parameter in rainwater samples (cations, anions, TDS, in mg/L, and EC in µS/cm)

sampling Sites	**F^-**	**Ca^{2+}**	**pH**	**EC**	**TDS**	**ZONE**
1	0.12	1.99	6.4	11.02	5.398	1
2	0.3	0.49	6.7	6.76	3.333	1
3	0.5	0.82	7.1	7.12	3.745	1
4	0.02	2.24	6.9	25.19	12.32	1
5	BDL	0.163	6.2	5.08	2.46	2
6	0.06	0.547	7.2	5.1	2.415	1
7	0.12	1.33	6.8	14.95	7.322	1
8	BDL	8.41	6.5	35.6	17.57	2
9	0.02	2.29	6.5	18.45	8.739	2
10	0.12	1.022	6.1	14.42	7.085	2
11	0.08	0.973	6.7	7.47	7.47	1
12	0.08	2.098	7	12.57	5.838	1
13	0.09	4.38	6.5	19.88	9.69	1
14	0.07	2.1	5.8	5.87	2.865	2
15	0.1	0.569	7.1	5.96	1.68	2
16	0.02	7.4	6.4	9.08	4.652	2
17	0.17	1.74	6	19.61	9.65	2
18	0.03	1.139	6.7	12.49	12.49	1
19	0.05	4.014	7.7	33	16.11	1
20	0.02	3.47	7.1	17.18	8.932	1
21	0.01	1.59	6.9	12.07	5.918	1
22	0.25	6.3	6.2	20.16	20.16	2
23	BDL	24.331	6.4	9.08	4.652	1
24	BDL	1.374	6.1	8.65	4.259	2

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ARTICLE

Application of Water Quality Index (WQI) and Regression Analysis of Groundwater in Budigumma Village, Anantapur District, Andhra Pradesh

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ABSTRACT

The research work is aimed at assessing the subsurface or groundwater suitability for human use or consumption depends upon the calculated water quality index values, correlation coefficient and regression analysis. The water quality index (WQI) is main important tool to calculate the characteristics of drinking water quality in rural, urban and industrial area. Different parameters which is measured and determination of the water quality index for selecting parameters. Further to study the correlation and regression method in this research work. Totally fifteen groundwater samples were collected from the Budigumma Village Anantapur district in the state Andhra Pradesh in India. Nine water quality parameters has been considered for the computation of water quality index such as pH, total dissolved solid (TDS), total hardness (TH), calcium (Ca), magnesium (Mg), nitrates (NO3), chlorides (Cl), sulphates (SO4), fluorides (F). The World Health Organization (WHO) has been assessed to the suitability of groundwater for drinking purposes or other uses for public and determining of WQI. This WQI index values ranged from 97.78 to 108.37. The study shows that 87% area comes under the poor category of drinking purposes and the remaining 13% comes under as good water for drinking purposes as per the WQI classification. The correlation and regression analysis gives as an outstanding device for the calculation of different parameter values within realistic degree of precision. The subsistence of strong correlation or relationship between the total hardness and magnesium is determined. The analysis of selected parameters revealed that proper treatment before use or consumption and protected from more contamination.

1. Introduction

In the world, groundwater is main basis of water supply using for human consumption. Groundwater occurs about all over the place under the aquifer [1]. Groundwater is a fixed resource and it is an infrequent advantage in various parts of the world. The water is a limited supply in the countries where the opposition is unrestrained among industry, domestic use and agriculture [2].

The quality of groundwater has develop into main water resource subject to quick raise in population, rapid built-up development, increasing petroleum and mining

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operations and as well use of pesticides and fertilizers in agriculture [3]. In India, many population use subsurface water as its main basis of drinking water [4]. Hence it develops into very significant to frequently verify the quality of groundwater to protect it.

Water Quality Index (WQI) is main important tools to converse in sequence on the water quality to the disturbed people and strategist [5]. This WQI is a arithmetical formula used to change into various great number of quality of groundwater data into a particular quantity or number. The study of groundwater quality in the research area has been calculated for the various groundwater samples using the method of weighted arithmetic index [6].

The study of the research area mainly deals with the WQI and analysis of correlation and regression method in Budigumma Village, Anantapur district in the state Andhra Pradesh, India.

This is main problem to create the environmental perception of a situation or fact among the peoples.

2. Materials and Methods

Totally fifteen groundwater samples were collected in the well sample station in Budigumma Village, Anantapur district in the state Andhra Pradesh. As per rule of standard, collection of the groundwater samples were analysed by physico - chemical characteristics of nine parameters such as pH, total dissolved solid (TDS), total hardness (TH), calcium (Ca), nitrates (NO_3), magnesium (Mg), chlorides (Cl), sulphates (SO_4), fluorides (F). These parameters were determined as per standard as presented in Table 1. Different instruments used for different parameters were accurate or calibrated prior to make use of detect readings[7]. Concordant readings for different parameters were to make certain accuracy and precision of results.

2.1 Water Quality Index (WQI)

Determination of Water Quality Index (WQI) is the following steps.

(1) Each nine parameters has been allocated a weight (wi) for overall water quality [7].

(2) Nitrate and Fluoride parameter has been allocated to the weight of 5 and 4 [8].

(3) Relative weight is calculated by weight of each parameter divided by summation of weight of each parameter. Nine parameters value as presented in Table 2.

(4) Quality rating scale (qi) is calculated by concentration (C_i) of each parameter and divided by standard values (S_i) [9]. The result values of each parameter is multiplied by hundred [10].

(5) Sub Index of ith parameter is calculated by relative weight and quality rate [11] as shown in Table 3.

(6) Water Quality Index values are determined and classified into excellent water (<50), good water (50 -100), poor water(100-200), very poor water(200-300), water unsuitable for drinking(>300) [12,13] as presented in Table 4.

2.2 Correlation Coefficient (r)

(1) Let independent parameter(x) and dependent parameter (y) are the variables.

(2) X_i and Y_i be n pairs of experimental or observed values of x any y variables (i =1, 2, 3……………n).

(3) Calculate the correlation coefficient (r) between the independent parameter(x) and dependent parameter (y) variables using SPSS 18 software [13, 14].

(4) Observed values of all parameters (a and b) were calculated from the software.

2.3 Regression Equation

The correlation study among different quality of water parameters, the regression analysis found out using software SPSS 18 [14]. The regression equation (y = a x + b) was used as a arithmetical or mathematical device in order to calculate different dependent parameters (y) of water quality by alternating the values for the independent parameters (x) and also a and b are constant variables.

Table 1. Statistics value of the analytical result of the parameters

Sl.No.	Water Quality Chemical Parameter	Minimum reading	Maximum reading	Mean value	Value of Standard. Deviation	Number of samples
1	Total Dissolved Solids (TDS) in mg/l	1015	1200	1107.67	57.535	15
2	pH	7.6	7.9	7.753	0.1060	15
3	Total Alkalinity (TA) in mg/l	60	550	371.33	102.878	15
4	Total Hardness (TH) in mg/l	320	440	397.33	41.312	15
5	Calcium (Ca) in mg/l	80	120	96.00	20.284	15
6	Magnesium (Mg) in mg/l	34	46	41.73	4.131	15
7	Nitrate (NO_3) mg/l	26	28	27.40	.632	15
8	Chloride (Cl) in mg/l	240	320	264.00	29.472	15
9	Fluoride (F) in mg/l	1.10	1.20	1.153	.0516	15
11	Sulphate (SO_4) in mg/l	72	86	76.53	4.502	15

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3. Results and Discussion

The results revealed that the study area which is based on water must be odourless, colourless and no turbidity. The analysis of physical characteristics of groundwater has turbidity as 2 NTU and iron as 0.02 mg/l present in fifteen samples. Statistics of the analytical results of groundwater water samples as shown in Table 1. The results shown that measured parameters at various points is not excessively high and among the measured values of these parameters at different locations is not excessively high and distinction range is very fine. The characteristics tests were determined make use of standard systematic methods, so as to minimize the determinate errors.

3.1. Water Quality Index (WQI)

World Health Organization (WHO) Indian standards, assigned weight (wi) and calculated relative weight (Wi) of nine parameters as shown in Table 2.

Table 2. WHO standards, assigned weight (wi) and calculated relative weight (Wi) of each parameter

Parameters	WHO -Indian Standard	Weight (wi)	Relative Weight (Wi)
pH	6.5 - 8.5	4.0	0.1333
TDS	500-2000	4.0	0.1333
TH	300-600	2.0	0.0667
Ca	75-200	2.0	0.0667
Mg	30-100	2.0	0.0667
Nitrate	1 - 45	5.0	0.1667
Chloride	250-1000	3.0	0.1000
Flouride	1-1.5	4.0	0.1333
Sulphate	200-400	4.0	0.1333
Total		**30.0**	**1.0000**

Table 3. WQI values for fifteen samples

Well No.	1	2	3	4	5	6	7	8
WQI	107.8	108.4	104.8	101.8	97.8	103.8	108.1	100.7
Well No.	9	10	11	12	13	14	15	
WQI	112.5	105.4	102.1	103.7	106.1	104.4	98.8	

Table 4. Classify Water quality based on the range of WQI value

Range of WQI Value	Classification of Water quality	Well Number	Percentages of Well Water Samples of nine parameters
<50.0	Excellent	Nil	0
50.0 -100.0	Good	5,15	13
100.0 – 200.0	Poor	1,2,3,4,6,7,8,9,10, 11,12,13,14	87
200.0 – 300.0	Very poor	Nil	0
>300.0	Unfit for use	Nil	0

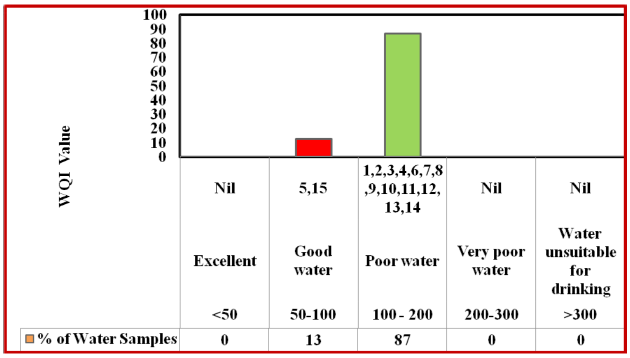

Figure 1. Percentage of well sampling points

The results revealed that, totally nine parameters contain pH, total hardness (TH), calcium (Ca), total dissolved solid (TDS), magnesium (Mg), nitrates (NO_3), chlorides (Cl), sulphates (SO_4), fluorides (F) has been used by WQI. The range of WQI calculated from (97.79-108.37) as presented in Table 3. Classification of water quality based on the range of WQI value as excellent water has less than 50.0. WQI values for the well number as zero. Good water quality has been WQI values in the range of 50.0 – 100.0 for the well number 5 and 15 as 13% of water samples as presented in Table 4. Poor water has water quality index (WQI) values ranged from 100.0 – 200.0 for the various well number 1, 2, 3, 4, 6, 7, 8, 9, 10, 11, 12, 13 and 14 as 87% of water samples are present as shown in Figure 1. Very poor quality have WQI values ranged from 200.0 – 300.0 for the well number as zero. The water is unfit for usage has WQI values greater than 300.0 for well as zero.

3.2 Correlation Analysis

The result of the correlation analysis revealed that Total hardness (TH) has positive and significant correlation or relationship with Magnesium (Mg) has value of 1.000 and also week correlation with calcium, nitrate, chloride, fluoride and then negative correlation with sulphate. TDS has week correlation with pH, Calcium, Sulphate and then negative correlation with Total alkalinity, Total Hard-

Table 5. Correlation values of ten parameters

Parameters	TDS	pH	TA	TH	Ca	Mg	Nitrate	Cl	F	SO_4
TDS	1									
pH	0.297	1								
Total Alkalinity	-0.467	-0.302	1							
Total Hardness	-0.033	-0.226	-0.268	1						
Calcium	0.132	-0.027	-0.285	0.327	1					
Magnesium	-0.033	-0.226	-0.268	1.000	0.327	1				
Nitrate	-0.159	0.085	0.024	0.044	-0.089	0.044	1			
Chloride	-0.032	-0.073	0.168	0.150	0.459	0.150	0.368	1		
Fluoride	-0.232	0.096	-0.162	0.205	-0.327	0.205	0.394	0.038	1	
Sulphate	0.215	0.056	-0.261	-0.391	0.025	-0.391	-0.331	-0.276	-0.254	1

ness, Magnesium, Nitrate and Chloride. pH has week correlation or relationship with Nitrate and Fluoride then negative correlation or relationship with Calcium(Ca), Sulphate (SO_4), Total alkalinity (TA), Total Hardness (TH), Magnesium (Mg) , Chloride (Cl) and Fluoride. Total alkalinity has week correlation or relationship with Nitrate (NO_3) and Chloride, then negative correlation or relationship with Calcium, Sulphate, Total Hardness, Magnesium and Fluoride. Calcium has week correlation or relationship with Magnesium, Sulphate and Chloride, then negative correlation or relationship with Nitrate and Fluoride. Magnesium has week correlation or relationship with Nitrate, Fluoride and Chloride, then negative correlation or relationship with Sulphate. Nitrate has week correlation or relationship with Fluoride and Chloride, then negative correlation or relationship with Sulphate. Chloride has week correlation with Fluoride then negative correlation with Sulphate. Fluoride has negative correlation or relationship with Sulphate. Different parameter has positive (+ve), negative (-ve) and week correlation or relationship as shown in Table 5. The regression equation for dependent and independent variables and R^2 values are shown in the Table 6.

3.3 Regression Equation or Analysis

The regression equation or analysis revealed that pairs of parameters such as independent and dependent variables or parameters establish to have improved and higher level of important or significance in their correlation or relationship coefficient are studied below. The regression equations for some pairs of parameters as shown in Table 4. Linear Plot between pairs of parameters as shown in Figure 2 to 6.

Table 6. Pairs of parameters (x and y) and regression equation with R^2 value

X and Y Parameters (y - dependent; x – independent)	**Regression Analysis or Equation**	R^2
TDS (y) –TH(x)	y = -0.045x + 1125	0.001
TDS (y) -Cl^- (x)	y = -0.062x + 1124	0.001
TDS (y) - SO_4^{2-}(x)	y = 2.744x + 897.6	0.046
TDS (y) - Ca^{2+}(x)	y = 0.375x + 1071	0.017
TDS (y) - Mg^{2+}(x)	y = -0.457x + 1126	0.001
TH (y) - Ca^{2+}(x)	y = 0.666x + 333.3	0.107
TH (y) - Mg^{2+}(x)	y = 10x – 20	1.000
Ca (y) - Cl^-(x)	y = 0.315x + 12.63	0.210
Ca (y) - SO_4^{2-}(x)	y = 0.112x + 87.36	0.000
Mg (y) - Cl^-(x)	y = 0.021x + 36.17	0.022
Mg (y) - SO_4^{2-}(x)	y = -0.359x + 69.21	0.153

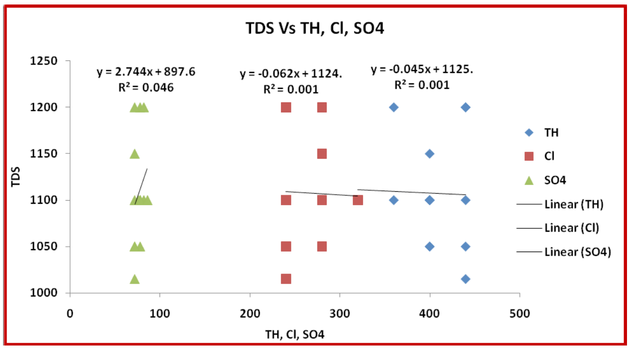

Figure 2. Linear Plot between TDS Vs TH, Cl and SO_4

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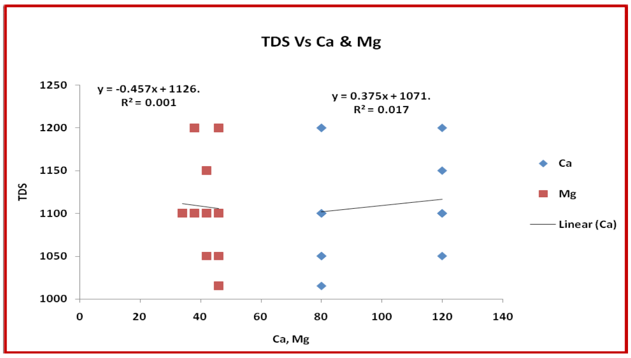

Figure 3. Linear equation plot between TDS Vs Calcium and Magnesium

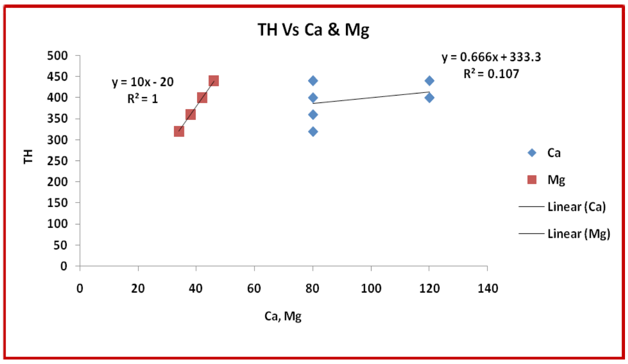

Figure 4. Linear equation plot between Total Hardness Vs Calcium and Magnesium

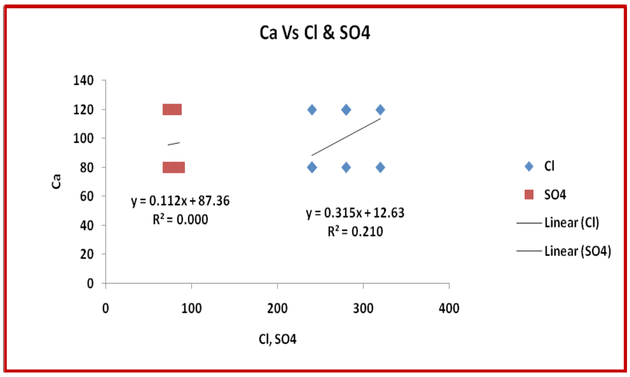

Figure 5. Linear equation plot between Ca Vs TH, Cl and SO_4

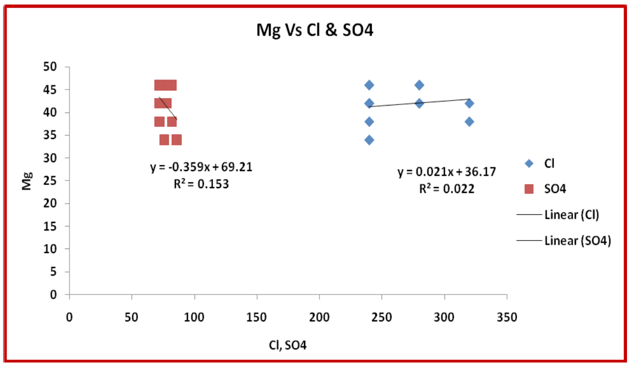

Figure 6. Linear equation plot between Mg Vs Cl and SO_4

4. Conclusions

The well water samples collected from the various sampling points in Budigumma village was analyzed. The study of experimental analysis on well water quality using nine physical and chemical characteristics indicate that check the water may be good and poor. The study revealed that the range of WQI values from 97.79 - 108.37. The Percentages of Well Water Samples of nine parameters were determined that the maximum in thirteen sample points. The WQI values of the study area has higher value which indicates that been indicated that deteriorate or depreciate water quality. The result of the correlation and regression analysis that Total hardness has strong positive correlated with magnesium. The remaining parameters are weak correlated and negative correlation with some others parameters. The physical and chemical analysis shows that the well water of the study area requires certain measure of treatment prior to consumption or utilization, and it also requires to be protected since the risks of contamination. The research study helps us to know the quality of the water and also to extend suitable management practices to protect or shelter the groundwater resources.

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