Reduction of Salinity of Water Using Acrylamide-Based Polyampholyte

Elvis Fosso-Kankeu*, Hasan Varachia and Frans Waanders

***Abstract*—Polyampholytes are charged macromolecules, which carry both acidic and basic groups. These groups dissociate, leaving ions on chains and counter ions in solution, under appropriate conditions, such as in aqueous solutions. After ionization occurs, there are positively and negatively charged groups on the polymer chain. Polyampholytes are able to absorb high dosages of anions and cations adequately and would ultimately be the best alternative for the reduction of salinity in water. In this study polyampholites with different ratio of cationic groups were synthesized and tested for the removal of sulphate from solution.**

High cationic Polyampholytes showed great sulphate absorption capacities and could remove 100% of sulphate content. These Polyampholytes are able to function on a relatively low dosage and over a short period of time. Polyampholytes may be a good solution towards the implementation of desalination plants.

***Keywords*—** absorption, desalination, polyampholyte, Salinity.

I. INTRODUCTION

Water scarcity predictions estimate that by the year 2025 about 50% of the world's population will suffer from water scarcity, approximating 3.5 billion people. Furthermore, South Africa will however exceed the accessibility of economically consumable drinking water by or before 2025. The immensely enduring trend of the growth of the population will inevitably place a further burden and strain on the requisite to clean water supplies, until or unless corrective mechanisms and techniques are established. [1]

The combustion of fossil fuel and metallurgical roasting processes release sulphates into the atmosphere as sulphur dioxide. Sulphur dioxide is converted to dilute sulphuric acid and by precipitation falls as acid rain contaminating surface water and resulting in high concentrations of sulphate in water. [2]

Mines contribute immensely to water pollution and are the main source of sulphate addition to water. Chemical pollution is partly due to the dispersion of mine waste in the environment. Coal preparation, acid mine drainage effluents and other anthropogenic methods and activities mostly contain elevated concentration of calcium, magnesium, sulphate and sodium. [3] Desalination is becoming a realistic alternative for coastal users as technology improves. [1] With only 3% of all available water on the planet being fresh water, seawater is the most abundant available source of drinking water and water for industrial use in many regions, and reverse osmosis is the dominant technology for production of drinking water. Seawater has a relatively high salinity, which sets a limit for the recovery by reverse osmosis at 30-50%. [4] This fundamentally leads to the need for improved salinity reduction techniques.

The drinking water from natural sources in coastal Bangladesh has become contaminated by varying degrees of salinity due to saltwater intrusion from rising sea levels, cyclone and storm surges, and upstream withdrawal of freshwater. The average estimated sodium intakes from drinking water ranged from 5 to 16 g/day in the dry season, compared with 0.6–1.2 g/day in the rainy season. This resulted in the annual hospital prevalence of hypertension in pregnancy being higher in the dry season. [5]

Guidelines for dietary salt intake have been established by the World Health Organization (WHO), but no guidelines have been released for safe salinity levels in drinking water, except that sodium levels > 0.2 g/L are unacceptable to taste. [6] High salinity levels in drinking water may have numerous direct and indirect impacts on health. In 2002 the WHO recognized health impacts of consumption of highly saline waters as a priority for investigation under its public health initiatives. [7].

High salinity levels have caused negative effects on plant growth globally. There exists a large influence of plant/crop damages and excessive salinity inhibits the growth of crop plants which include osmotic effects, as well as the direct toxicity caused by certain ions. [8]

Common practices for the removal of sulphate and calcium mainly consist of the use of coagulation/flocculation processes or reverse osmosis, which are either ineffective, costly or result in toxic by-products. Adsorption is a better alternative to remove such pollutants from water, giving the opportunity to recover and control the pollutant while regenerating the adsorbent. [9, 10-21]

Polyampholyte hydrogels (PAHs), which contain ionic components, otherwise known as ionic side groups, are

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commonly referred to as smart hydrogels, because of the reversible deformations. These could be swelling, bending, rotating or translating, which occurs when an external stimulus is applied. The repeating units of PAHs, which are positively and negatively charged, are the most suitable functional groups for the adsorption of various toxic ionic particles.

II. EXPERIMENTAL

Polyampholytes are synthesized hydrogels which were designed by radical polymerization of monomers at room temperature and allowed to bind. A cation and anion were used in combination with a crosslinking agent, an accelerator and an initiator. Different parameters namely, the molar ratio of cation to anion, dosage and time used to determine optimum parameters in a synthetic solution based on the absorption capacity of Sulphur. Seawater and the Potchefstroom municipality tap water were considered as polluted water sources to be treated using the synthesized polyampholytes. The various polyampholytes were synthetized by adapting a method previously reported [9, 22].

A. Materials

The following materials were used and were purchased from Rochell, SIGMA, ACE and Merck in South Africa.

i. 3-acrylamidopropyl-trimethylammonium-ATP as a cation.
ii. Acrylic acid-Aac as an anion.
iii. N,N'methylenebis-acrylamide (MBA) as a cross linker.
iv. Potassium persulfate (KPS) as an initiator.
v. N,N,N,N'-tetramethylethylenediamine (TEMED) as the accelerator.
vi. For the preparation of a synthetic solution: Sodium sulphate anhydrous
vii. Seawater was couriered from Cape Town.
viii. Water was taken from the tap for Potchefstroom municipality water.

B. Synthesis Of Hydrogel And Synthetic Solution

The synthesis of the polyampholyte was carried out at room temperature. Before the process was initiated, potassium persulfate (KPS) was diluted in deionized water (DW) to achieve a liquid solution of 2% (w/v) and hence 2 g was diluted into 100ml of DW.

Five different cation to anion molar ratios were synthesized (3:1.7, 2.4:2.3, 1:1, 1.8:2.8, 1.2:3.4). In a 250 mL beaker, 0.28 mg MBA was diluted with 6 mL of DW and mixed with a magnetic stirrer to ensure the complete mixing and dissolving of the MBA. By means of a pipette the different volumes of cation (ATP) and anion (Aac) were added, based on the different ratios as indicated in sTable 1.

TABLE I
POLYAMPHOLYTE HYDROGEL CALCULATED VOLUMES BASED ON MOLAR RATIOS

Polymer name	Cation ratio	Anion ratio	ATP volume [ml]	Aac volume [ml]
A	3	1.7	4.856	0.760
B	2.4	2.3	4.440	1.176
C	1	1	4.400	1.216
D	1.8	2.8	3.928	1.688
E	1.2	3.4	3.150	2.466

The initiator (KPS) of volume 3.2 mL and the accelerator (TEMED) of volume 0.5 mL were added to the solution. The solution was mixed for approximately five minutes on a magnetic stirrer and the magnetic stirrer was removed and the beaker with the solution was placed in the incubator for a duration of 24 hours at 25^{o}C and 90 rpm, which allows for the total formation of a hydrogel. This process was repeated for the different molar ratios referred as polymer A-E as indicated in Table 1. Fig 1. bellow illustrates the chemical interaction for preparation of the polyampholytes.

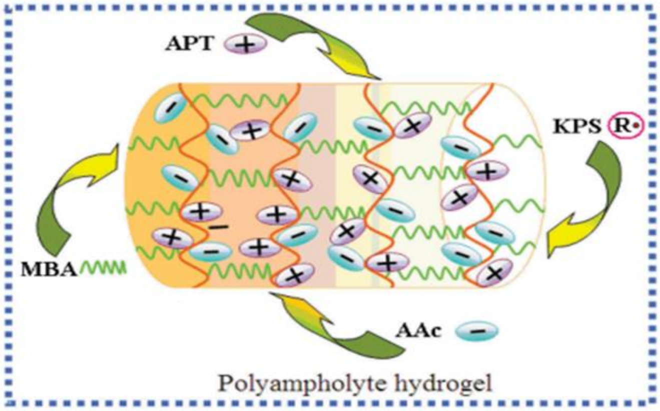

Fig 1

Fig1. Represents The Polyampholyte hydrogel configuration with the cross-linker taken from Kudaibergenov & Sigitov [22] The formed polyampholytes hydrogels, after 24 hours of reaction time, were then cut up into +- 4-5 mm cubes and deionized water (DW) was added to the beakers and stored for 24h at room temperature. This allowed for the dilution of the unreacted species in the DW. The hydrogels and DW mixture were then filtered after the 24 hours and the process for the removal of unreacted species was repeated for a duration of seven days.

On the seventh day of filtration the wet hydrogels were then placed in the incubator for drying. The hydrogels were dried for 24 hours at 40^{o}C. These polyampholyte hydrogels were then crushed by a hand stone crusher to form a powder.

A 1 L base synthetic solution of 1000 ppm sulphate was made by dissolving Sodium sulphate anhydrous into 1 L DW. For the adsorption experiments, working solutions of 50 ppm concentration were prepared.

C. Characterization Of Hydrogel

The powdered hydrogels were sent for analyses using Fourier-transform infrared spectroscopy (FTIR) and Scanning Electron Microscopy with Energy Dispersive X-Ray Spectrometer (SEM/EDS).

D. Absorption experiments

To determine the best polyampholyte ratio for the removal of sulphate from solution, 0.3 g of each molar ratio was dosed into a 100 mL of synthetic solution in a 250 mL Erlenmeyer flask at room temperature of 25^{o}C for a duration of 1 hour in an incubator at 160 rpm and the optimum ratio was evaluated. Similarly, various masses (0.1 g, 0.2 g, 0.3 g, 0.4 g) of the optimum ratio were dosed into a 100mL of synthetic solution in a 250 mL Erlenmeyer flask at room temperature of 25^{o}C for a duration of 1 hour in an incubator at 160 rpm and the optimum dosage was evaluated. In a likewise fashion the optimum contact time was determined considering sulphate exposure to the polyampholytes for the following time intervals, 25, 50, 75, 100, 125 mins. Each of these were based on the turbidimetric sulphate results and were centrifuged prior to test. All tests were conducted in duplicate.

E. Experimental Analysis And Models

The sulphate percentage removals were determined using the following formula:

$$\% \, Absorption = \frac{C_o - C_e}{C_o} \; X \; 100$$

Where C_o is the initial concentration in mg/L of sulphate and C_e is the average equilibrium concentration after dosage.
The two kinetic models used were the pseudo-first-order kinetic model and the pseudo-second-order kinetic model.

The pseudo-first-order kinetic model:

$$\log(q_e - q_t) = \log(q_e) - k_1 \frac{t}{2.303}$$

The pseudo-second-order kinetic model:

$$\frac{t}{q_t} = \frac{1}{(k_2 \, q_e^2)} + \frac{1}{q_e} t$$

With q given as:

$$q_t = \frac{(C_o - C_t)V}{m}$$

V is the volume in liters, m is the mass in mg and q is dimensionless. The two constant achieved are K_1 and K_2 based on the linear regression and can be used in further approximations.

III. RESULTS AND DISCUSSION

A. FTIR Analysis

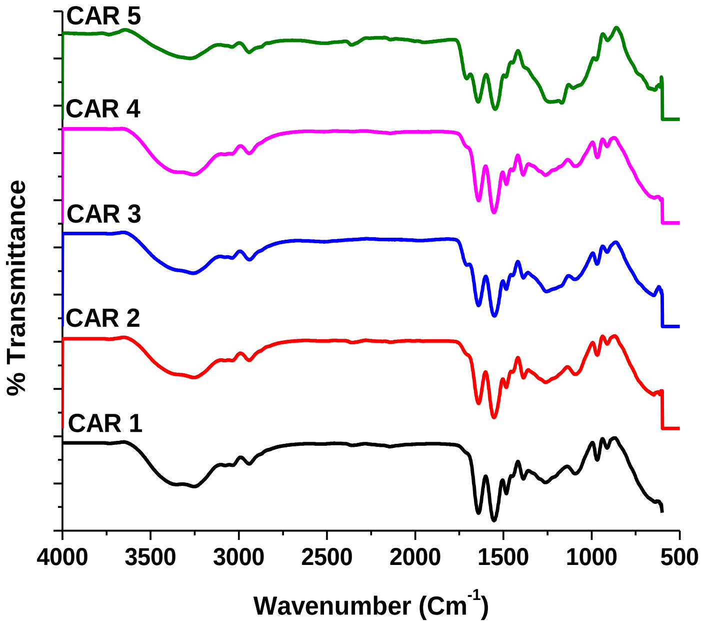

Fig. 2

Fig 2. Represents the FTIR functional group graph characterization results obtained. The synthesis of the five absorbents of different cationic and anionic ratios have shown a common trend in peaks indicating consistency in its functional groups. The different ratios are shown with CAR 1 being most cationic and CAR 5 being the most anionic ratio The peaks indicate good absorption capacities. In the range 3700-3200 cm^{-1} a clear broad O-H peak is observed between 3650-3200 cm^{-1} showing the presence of the alcohol functional group. Amine and amide functional groups of N-H bonds maybe present within the 3500-3300 cm^{-1}. Within the range 3200-2700 cm^{-1} the presence of a small peak may correspond to alkane, aryl, vinyl and carboxylic acid O-H bonds functional groups. In the range 1680-1450 cm^{-1} wavenumbers, there are steepest peaks with a clear distinction which can be ascribed to C-C=C symmetric stretch and C=O stretch functional groups which have high adsorption affinity. [23]

B. SEM Analysis

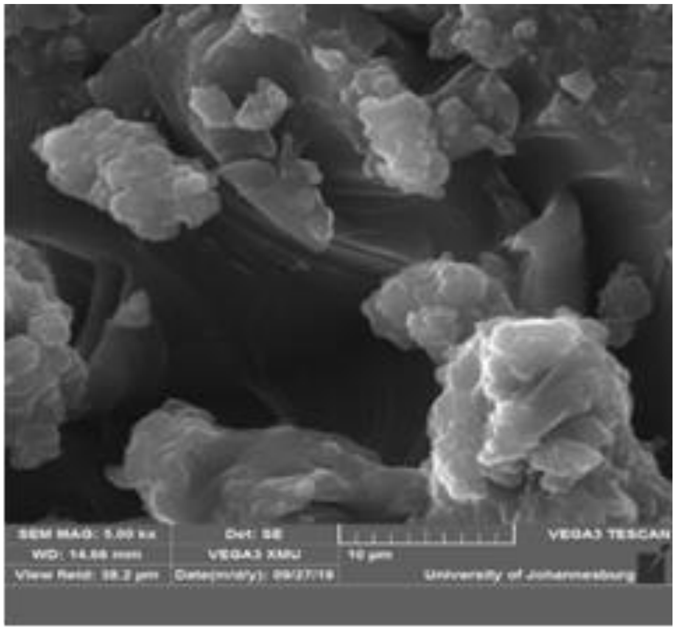

Fig 3 (a)

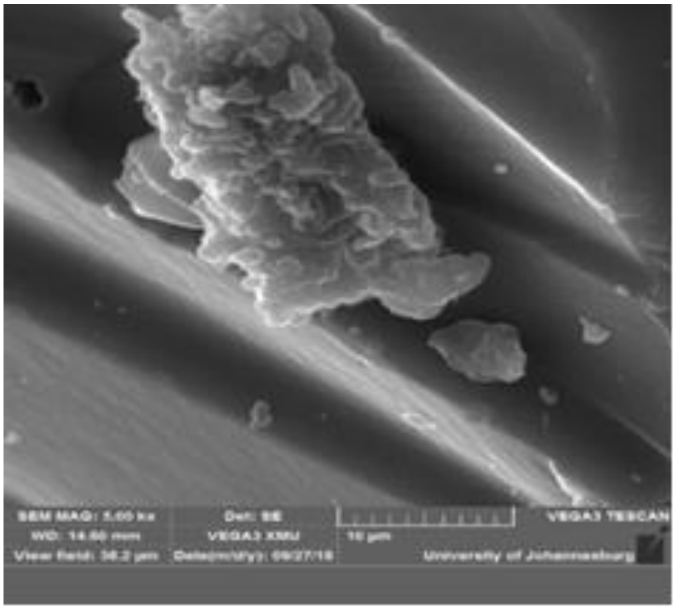

Fig 3 (b)

Fig.3 indicates two absorbents SEM analysis with (a) being the most cationic absorbent and (b) being the most anionic absorbent.

A clear difference in the morphology of the polymers can be observed as the most cationic polyampholyte exhibits a rough surface while the most anionic polyampholyte exhibits a more smooth surface The difference in morphology can be ascribed to the variation in the ratio of the ingredients that certainly affected the chemical reaction; however, there was not major difference likely to significantly affect the adsorption capacity.

C. EDS Analysis

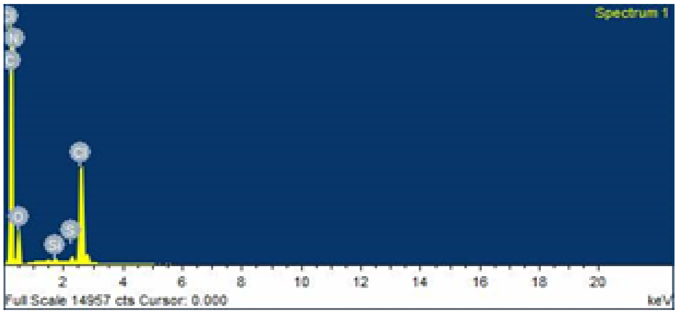

Fig 4 (a)

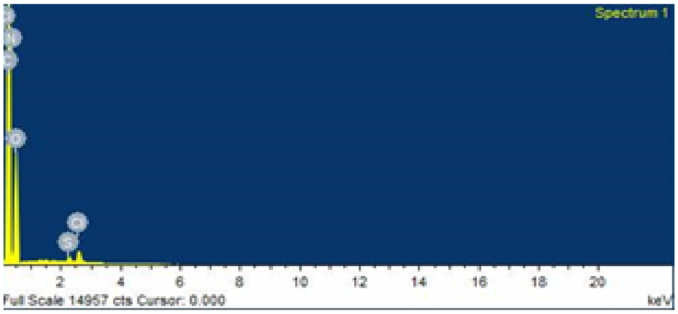

Fig 2 (b)

Fig. 4 indicates two absorbents EDS spectra analysis with (a) being the most cationic absorbent and (b) being the most anionic absorbent.

A consistency in the elements present are noted as expected with different peak levels indicating that the desired output based on the ions expected for cationic and anionic compounds and hence illustrating the different range in charges. An energy total level ranged between 6-7 keV which is suitable for absorption. The energy level per compound is also consistent whereby at around 2.5 keV, Cl^- is observed and at around 2.3 keV, S^{2-} can be seen. The steep Cl^- curve in (a) confirms the cationic capacity of (a) compared to (b).

D. Tubidimetric Sulphate Analysis

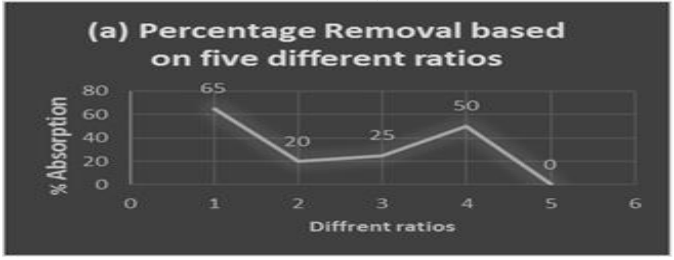

Fig 5 (a)

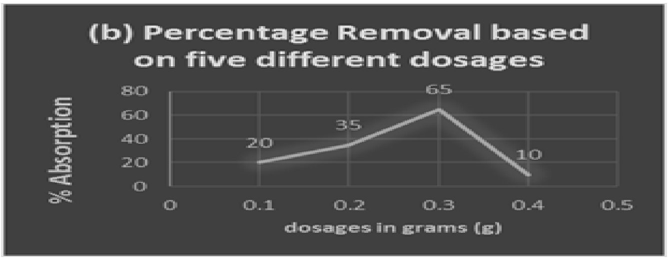

Fig 5 (b)

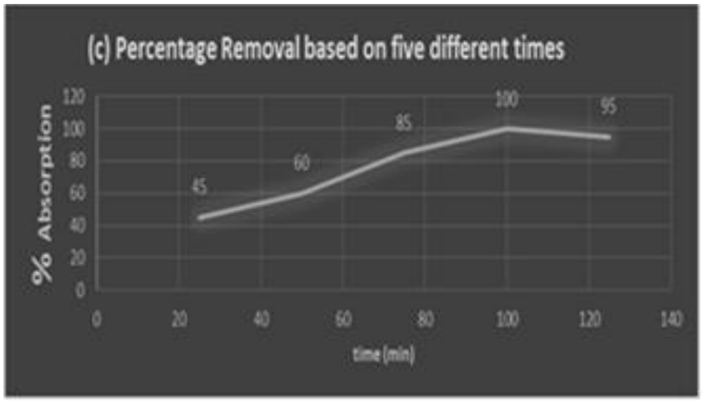

Fig 5 (c)

The Tubidimetric sulphate analysis for the five different cationic and anionic absorbents indicated a 65% removal of sulphate for ratio one being the most cationic compound with a ratio of 3:1.7 cation to anion as indicated in Fig. 3(a). The investigation of the effect of adsorbent dosage showed (Fig. 3(b)) a dosage of 0.3 g was sufficient to achieve a higher removal rate corresponding to 65% removal of sulphate. The investigation of the effect of time on the removal of sulphate showed that 1h40 mins exposure time was enough to achieve 100% removal of sulphate from solution.

E. Kinetic Models

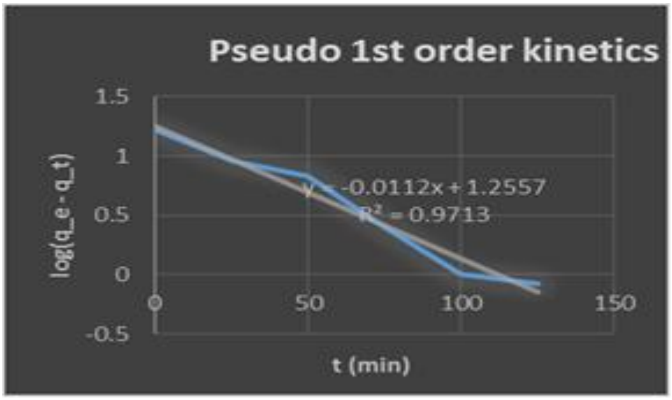

Fig 6 (a)

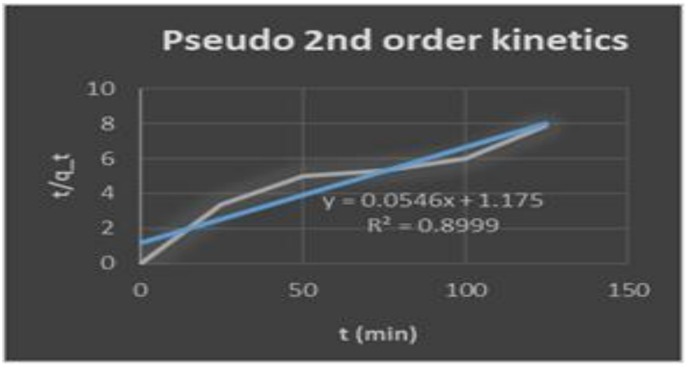

Fig 3 (b)

A close approximation of the kinetics models indicates a satisfactory trend line with a better fit for the Pseudo first order model having a regression of 0.97. The constant values for the models were calculated as k_1= 0.025794 and k_2=0.003064 for the pseudo first and second order kinetic model respectively. The relative fitness of the pseudo second order kinetic model to the adsorption data also shows that the adsorption of sulphate by the polyampholyte occurs through a combination of physical and chemical mechanisms as expected [24-36].

F. Practical Test Analysis

Fig 4 (a)

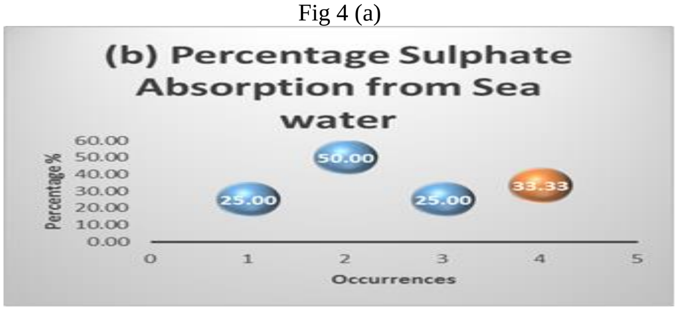

Fig 7 (b)

Good sulphate extraction capacities are observed and can be seen in Fig. 7 (a) and (b). Fig. 7(a) indicates absorption capacities based on samples of the Potchefstroom tap water which contained a sulphate content bellow the South African standards for acceptable water but a high content of 110 mg/L an average absorption capacity of 82.42% was achieved reducing the sulphate content to just above 10 mg/L. Fig. 7(b) indicates the rate of absorption of sulphate from Sea water samples containing 2000 mg/L of sulphate; the polyampholyte was in this occasion able remove 33.33% of sulphate from sea water which is however low but can be optimized further with a specific time and dosage analysis.

IV. CONCLUSION

The absorption capacities of Polyampholytes have shown phenomenal characteristics towards the reduction of sulphate content in water without being energy intensive. This can be seen as a best and optimal solution mechanism towards further development in the reduction salt from water. The cation and anion ratios facilitated the exchange of ions allowing for the absorption by ions. A more cationic hydrogel has exhibited encouraging adsorption capacity for the reduction of sulphate in water, this is due to the negative charge carried by sulphates. Further improvement of the polyampholyte is therefore required for application in the treatment of solutions with high salinity such as sea water.

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