

Study on the Effect of Different Electrode on Capacitive Deionization Microfluidic Desalination

J.T. Heng^{1,2*}, Hayder A. Abdulbari^{1,2}

¹Centre of Excellence for Advanced Research on Fluid Flow, Universiti Malaysia Pahang, 26300 Gambang, Pahang, Malaysia

²Faculty of Chemical & Natural Resources Engineering, Universiti Malaysia Pahang, 26300 Gambang, Pahang, Malaysia

*Corresponding author E-mail: h.jingthin@gmail.com

Abstract

Recent year, microfluidics desalination technology is on its immersing path which utilizes the domination of many apparent fluids physical properties (viscosity and surface tension) in the micro-flow systems. As compared to the traditional and commercially applied desalination methods, microfluidics overcomes most of the drawbacks such as high power consumption and low separation performance. It is believed that the flow of liquids in the micro-scaled designed structures will optimize the separation efficiency and will definitely lead to higher desalination performance. In the present work, a microfluidic desalination chip was introduced. The microfluidic desalination chip was fabricated using polydimethylsiloxane (PDMS) soft lithography method and two types of electrode were used which are titanium and aluminium. The desalination efficiency was being observed, analyzed and evaluated at the constant flow rate of 90 mL/h using capacitive deionization method. The desalination efficiency with titanium and aluminium electrode was achieved with 15% and 65%, respectively. The surface morphology of both used and unused electrodes was observed by using scanning electron microscopy (SEM). The findings in this work show that the desalination efficiency was rely on the electrode surface properties.

Keywords: *Capacitive deionization; desalination; electrode; aluminium; titanium.*

1. Introduction

Desalination is gaining more attention since it is the crucial technology in order to solve the water scarcity issue by effectively using the abundant natural sourcing (sea water). There are several traditional and commercialized desalination methods such as multi-stage flash distillation (MSF) [1–5], multi-effect distillation (MED) [1,6–9], electrodialysis (ED) [10–13] and reverse osmosis (RO) [14–18]. These available methods provide high productivity of potable water however there are several drawbacks such as high thermal energy was needed for the distillation based techniques and high power consumption needed for RO due to the fouling problem faced in each membrane technologies which reducing the overall effectiveness of the process [16,17,19–21].

Recently, the development of microfluidic technology has attracted the attention of researchers in science and engineering [22–28]. The technology has also grabbed the interest from those who are extensively working in the desalination technology since it created more opportunities for the researchers to overcome some drawbacks faced in macro technologies. Capacitive deionization (CD) is an ion electrosorption technology which is applied in microfluidic desalination work and was demonstrated by using capacitive wire-based technology to remove the sodium and chloride ion with a pair of porous carbon coated electrode [29]. CD technology is emerging and widely explored for water desalination application nowadays due to its simplicity for removal of charged ions from aqueous salt solution [30–32]. The attracted ions were being trapped inside the porous structure charged electrodes. The trapped ions were eventually reducing the effective surface area of the electrode causing the ion removal efficiency decreases with

time. A rinsing process was needed to be carried out in batches to detach the trapped ions from the electrodes and hence the desalination process was unable to be operated in continuous mode and eventually increase the overall processing time.

There are two most common electrode being used in microfluidic desalination which is silver/silver chloride and carbon electrode. Silver/silver chloride electrode was being used for the electrochemical desalination together with the Nafion membrane based on the Faradaic reaction in order for the sodium ion to be removed during the oxidation reaction [33]. 90% salt removal was demonstrated with a maximum flow rate of 40 μ L/min and initial salt solution concentration of 0.6 M NaCl. A supercapacitor was designed using capacitive deionization technique where two porous activated carbon electrodes were placed facing each other and 1 V potential was applied. The ions from the solution were moved towards the oppositely charged electrical double layer. It was reported that > 50% of charge efficiency was needed in order to achieve 70% desalination in this CD [34].

Aluminium is very favorable to be used in various fields such as food and construction industries due to its fully resistant properties to rust and highly against to weathering factor. Aluminium reacts spontaneously with water and air to become aluminium oxide due to its oxidation properties which form its corrosion protective layer. Despite its very high electrical conductivity ability, aluminium very seldom to be used as an electrode since it has a short lifespan in the electrolyte which contains water [35].

Titanium is favorable to be used in naval ships, missiles, aircraft, electric power plant, heat exchanger and desalination plants since it showed a remarkable corrosion resistivity towards salt water and almost completely unaffected by the marine environment [36–40]. Titanium has been widely used in desalination fields which in-

cludes magnetized titanium dioxide for sea water purification, titanium tubes in multi-stage flash (MSF) desalination and titanium oxide nanoparticles for enhancing nanofiltration desalination [3,36,41].

Despite the promising nature properties of aluminium and titanium as electrodes, there is still limited study in investigate the performance of these electrodes in microfluidic desalination. In this present study, the effect of the aluminium and titanium electrode on the conductivity value of the desalted outlets solution was being studied in order to determine the salt removal efficiency by the desalination chip. The surface morphology was also being observed under scanning electron m (SEM).

2. Material and methods

2.1. Chemicals

Sodium chloride (NaCl) powder, ethylene glycol, ammonium fluoride, aluminium foil and 200 μm titanium sheets were purchased from Sigma-Aldrich (St. Louis, Missouri, USA), SU-8 (GM1060 for layers between 5 and 27 μm , GM1070 for layers between 15 and 200 μm) from Gersteltec (Pully, Switzerland) and polydimethylsiloxane (PDMS) and its curing agent from Dow Corning (Michigan, USA).

2.2. Feed solution preparation

35 g/mL of NaCl solution which equivalent to 0.6 M NaCl was prepared by dilution method.

2.3. Wafer fabrication

Table 1 showed the details of the parameters for wafer fabrication. 5 mL of SU-8 was pipetted onto a clean wafer and spin coat at 900 rpm for 1 minute to obtain the desired thickness of 100 μm . The wafer was pre-baked at 65 $^{\circ}\text{C}$ for 15 minutes and increase to 95 $^{\circ}\text{C}$ for 2 hours. After pre-bake, the wafer was exposed under micro-pattern generator (μPG) to obtain the desired microchannel design.

The wafer was then post-bake at 65 $^{\circ}\text{C}$ for 15 minutes and increase to 95 $^{\circ}\text{C}$ for minutes. The steps were repeated for the second layer coating and exposed.

Table 1: Parameters for wafer fabrication

	Wafer layer	
	First layer	Second layer
Volume of SU-8 (mL)	5	5
Spin coat	900 rpm, 1 minute	900 rpm, 1 minute
Pre-bake	65 $^{\circ}\text{C}$, 15 minutes	65 $^{\circ}\text{C}$, 15 minutes
	95 $^{\circ}\text{C}$, 2 hours	95 $^{\circ}\text{C}$, 2 hours
Expose	Yes	Yes
Post-bake	65 $^{\circ}\text{C}$, 15 minutes	65 $^{\circ}\text{C}$, 15 minutes
	95 $^{\circ}\text{C}$, 40 minutes	95 $^{\circ}\text{C}$, minutes
Develop	No	Yes
Hard-bake	No	135 $^{\circ}\text{C}$, 2 hours

2.4. Desalination chip fabrication

Figure 1 showed the details of microfluidic desalination chip fabrication steps. The experiment was conducted in the PDMS based microfluidic model system. The PDMS was mixed with its curing agent at the ratio of 10:1 using a planetary centrifugal mixer (model: Thinky, USA). The well-mixed PDMS was poured onto the wafer which ready put in a petri dish. The PDMS was then being degassed in a desiccator for 1 hour in order to remove all the air bubbles trapped inside and then heated for 2 hours in the oven.

After heating, the first part of PDMS was cut using a scalpel and peeled off from the wafer with tweezers. The electrode was brought to be placed onto the wafer. The first part of PDMS and the second part of PDMS were brought to the plasma cleaner for surface treatment. Lastly, both treated PDMS were pasted together and pressed slightly. The entire fabrication process was done in the clean room to avoid any contamination on the chip. The steps were repeated to assemble the PDMS with the third part of PDMS. The desalination chip was fabricated by a combination of three layers of PDMS and two electrodes.

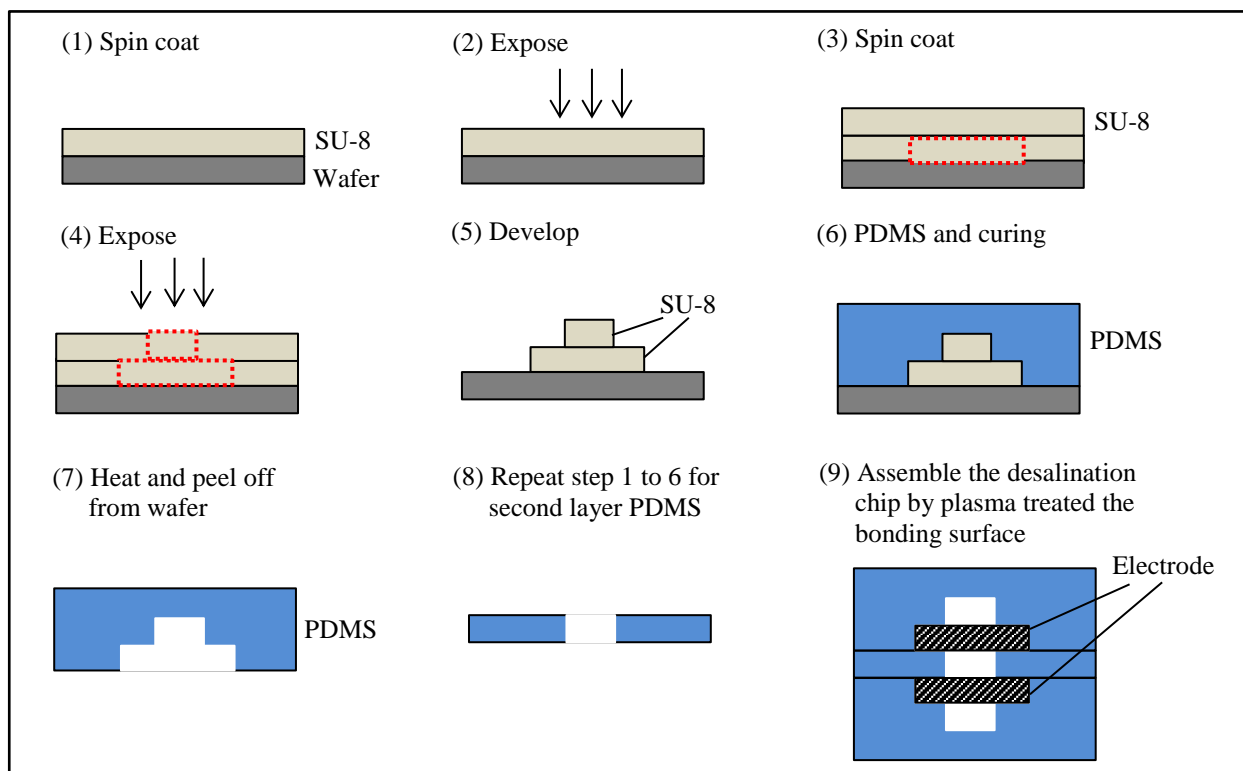


Fig. 1: Fabrication steps of the desalination chip.

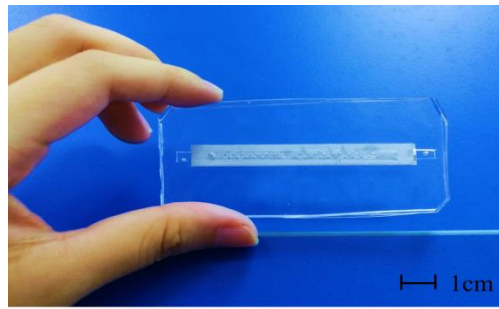


Fig. 2: Microfluidic desalination chip.

2.6. Experimental set up

Figure 3 showed the experimental set up. The negative pressure-driven flow with 90 mL/h was generated by microfluidic flow sensor (Elveflow OB1 MK3, France) and the constant voltage was applied by a programmable DC power supply (Rigol Technologies

Inc., USA). The desalination was conducted for 1 hour using both aluminium and titanium electrodes. The conductivity of outlet solution was measured using conductivity meter (Eutech PCD650, SG).

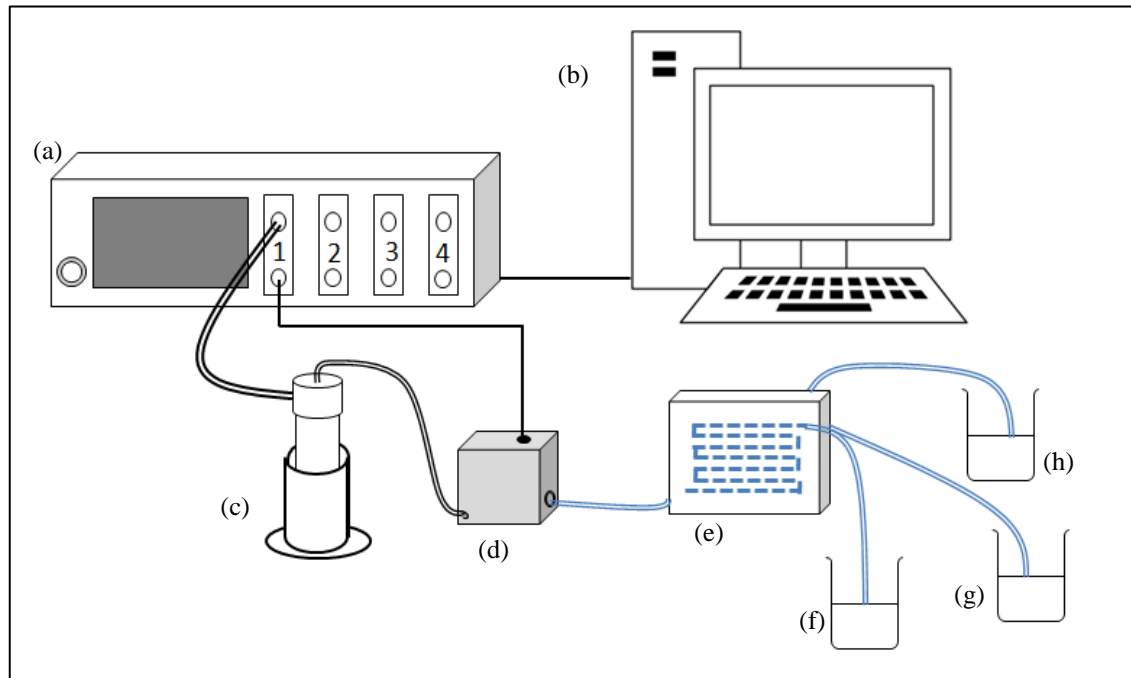


Fig. 3: Schematic diagram of experimental setup which consisted of (a) flow sensor, (b) computer, (c) feed reservoir, (d) pressure and vacuum controller, (e) desalination chip, (f) beaker for concentrated solution at anode side, (g) beaker for diluate and (h) beaker for concentrated solution at cathode side.

3. Result and discussion

3.1. Desalination efficiency

The experiment was conducted with two different electrodes which are aluminium and titanium. The desalination efficiency of the microfluidic desalination chip was measured with the conductivity of the desalted solution and the result versus time was plotted as shown in Figure 4. The trend of salt removal efficiency by the microfluidic desalination chips was observed for 1 hour. Both of the desalination chips took approximately 20 minutes to achieve to the maximum salt removal efficiency of 65% and 15% by utilizing aluminium and titanium electrodes, respectively. The titanium electrode desalination chip maintains almost constant salt removal efficiency since its surface resists to oxidation in salt water [36–40]. Aluminium electrode desalination chip showed very high potential in desalination however it encountered rapid

drop in salt removal efficiency after 20 minutes due to its surface oxidation in salt water.

Aluminium electrodes show higher salt removal efficiency but due to the rapid oxidation occur, the surface of the electrodes was treated by coating with nano-powder. The aluminium electrode was coated with carbon nano-powder which act as a conductive protective layer to avoid it from the oxidize problem. Figure 5 showed the salt removal efficiency performance by the coated aluminium desalination chip. The carbon nano-powder coating layer successfully slowed down the oxidation speed of aluminium where the salt removal efficiency was maintained at an average of approximately 55%.

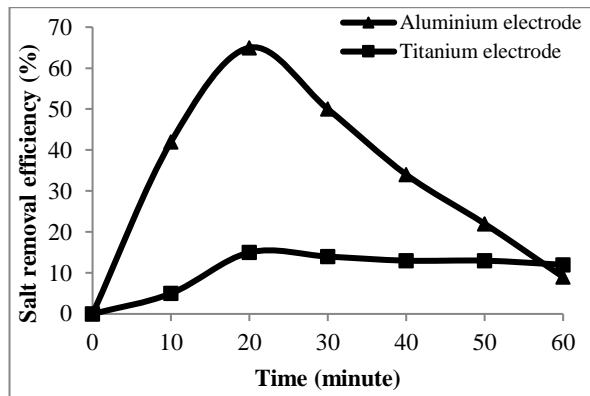


Fig. 4: Salt removal efficiency for different electrodes.

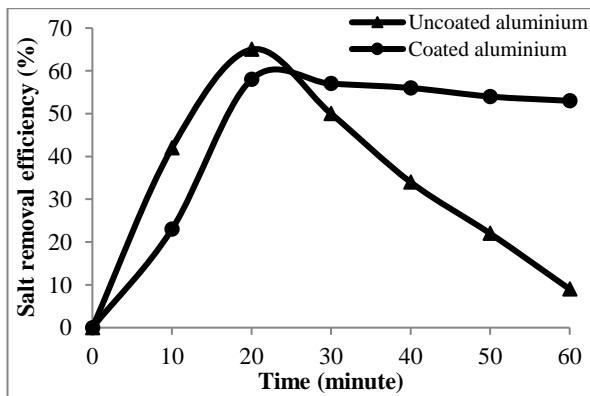


Fig. 5: Salt removal efficiency for uncoated and coated aluminium.

3.2. SEM Analysis

The SEM analysis was performed to observe the surface morphology of both titanium and aluminium electrode. Besides, the SEM image also being used to analyze and relate the performance of carbon coating on the aluminium. The surface morphology of uncoated and coated aluminium was observed before and after the desalination as shown in Figure 6. The SEM image of uncoated aluminium showed a very smooth surface before the experiment while a rough surface after the desalination since oxidation happened and oxide layer was formed. Figure 6 (c) showed the coated aluminium with a coarse surface which indicates the carbon nano-powder was evenly coated on the aluminium electrode. The damaging of the carbon coated layer was observed in Figure 6 (d) which was the SEM image captured at 30 minutes of the experiment. The surface morphology was not as clear and organized as Figure 6 (c) due to the high inlet flow rate causes parts of the coating layer was washed off. This was related to the result as shown in Figure 5 where the salt removal efficiency experienced slightly decreases after 30 minutes of the desalination experiment. The destructed carbon coating surface causes the aluminium was exposed to the salt water and oxidation occurred hence reduces the desalination efficiency.

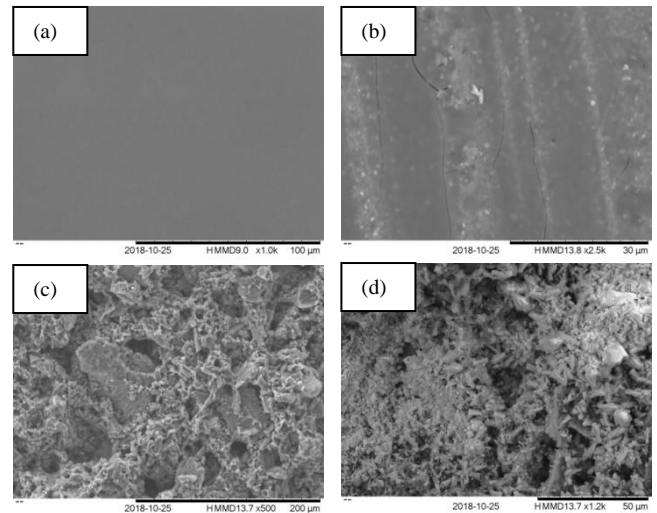


Fig. 6: Surface morphology of (a) uncoated aluminium, (b) uncoated aluminium at 30 minutes, (c) coated aluminium and (d) coated aluminium at 30 minutes.

4. Conclusion

In this work, the capacitive deionization microfluidic desalination efficiency was related to the oxidizing property of both electrodes which are titanium and aluminium. As noticed from the first part of experiment, the aluminium was highly potential to be used as the electrode for desalination however the oxidizing property causes the desalination efficiency reduced significantly over the time. The aluminium was then being coated using carbon nano-powder which serves as the protective layer to overcome the oxidizing problem of the electrode. Based on the results, the coated aluminium showed a better result where the oxidation has been prevented and the desalination efficiency was maintained at a nearly constant value which was approximately 55%.

Acknowledgement

The authors are grateful to Universiti Malaysia Pahang (PGRS180393) for financial assistance.

References

- [1] Roy Y, Thiel GP, Antar MA & Lienhard JH (2017), The effect of increased top brine temperature on the performance and design of OT-MSF using a case study. *Desalination* 412, 32–8.
- [2] Shams El, Din AM, El-Dahshan ME & Mohammed RA (2005), Scale formation in flash chambers of high-temperature MSF distillers. *Desalination* 177, 241–58.
- [3] Morris AR (1979), The use of titanium tubes in MSF desalination plants. *Desalination* 31, 387.
- [4] Baig H, Antar MA & Zubair SM (2010), Performance characteristics of a once-through multi-stage flash distillation process. *Desalination Water Treat* 13, 174–85.
- [5] Shams AM, Din E, El-Dahshan ME & Mohammed RA (2005), Scale formation in flash chambers of high-temperature MSF distillers. *Desalination* 177, 241–58.
- [6] Frantz C & Seifert B (2015), Thermal Analysis of a Multi Effect Distillation Plant Powered by a Solar Tower Plant. *Energy Procedia* 69, 1928–37.
- [7] Filippini G, Al-Obaidi M, Manenti F & Mujtaba IM (2018), Performance analysis of hybrid system of multi effect distillation and reverse osmosis for seawater desalination via modelling and simulation. *Desalination* 448, 21–35.
- [8] Ali MT, Fath HES & Armstrong PR (2011), A comprehensive techno-economical review of indirect solar desalination. *Renewable Sustain Energy Review* 15, 4187–99.
- [9] Khawaji AD, Kutubkhanah IK & Wie JM (2008), Advances in seawater desalination technologies. *Desalination* 221, 47–69.

- [10] Goodman NB, Taylor RJ, Xie Z, Gozukara Y & Clements A (2013), A feasibility study of municipal wastewater desalination using electro-dialysis reversal to provide recycled water for horticultural irrigation. *Desalination* 15, 77–83.
- [11] Gumuscu B, Haase AS, Benneker AM, Hempenius MA, van den Berg A & Lammertink RGH (2016), Desalination by Electro-dialysis Using a Stack of Patterned Ion-Selective Hydrogels on a Micro-fluidic Device. *Advanced Functional Materials* 26, 8685–93.
- [12] Deng D, Dydek EV, Han J-H, Schlumpberger S, Mani A & Zaltzman B (2013), Overlimiting Current and Shock Electrodialysis in Porous Media. *Langmuir* 29, 16167–77.
- [13] Kwon HJ, Kim B, Lim G & Han J, “A water permeable ion exchange membrane for desalination”, 19th International Conference on Miniaturized Systems for Chemistry and Life Sciences, (2015), pp:1202-1204.
- [14] Tang CY & Leckie JO (2007), Membrane Independent Limiting Flux for RO and NF Membranes Fouled by Humic Acid. *Environment Science Technology* 41, 4767–73.
- [15] Adham S, Hussain A, Matar JM, Dores R & Janson A (2013), Application of Membrane Distillation for desalting brines from thermal desalination plants. *Desalination* 314, 101–8.
- [16] She Q, Wang R, Fane AG & Tang CY (2016), Membrane fouling in osmotically driven membrane processes: A review. *Journal of Membrane Science* 499, 201–33.
- [17] Li D & Wang H (2010), Recent developments in reverse osmosis desalination membranes. *Journal of Material Chemistry* 20, 4551.
- [18] Pangarkar BL, Sane MG & Guddad M (2011), Reverse Osmosis and Membrane Distillation for Desalination of Groundwater: A Review. *ISRN Materials Science* 2011, 1–9.
- [19] Ruiz GA, Melián MN & Mena V (2018), Fouling characterization of RO membranes after 11 years of operation in a brackish water desalination plant. *Desalination* 430, 180–5.
- [20] Abdulrazaq JA (2014), Effect of the Scale Formation on the Performance of Recirculation MSF Plant. *Basrah Journal for Engineering Science* 41, 4141.
- [21] McGovern RK, Weiner AM, Sun L, Chambers CG, Zubair SM & Lienhard VJH (2014), On the cost of electro-dialysis for the desalination of high salinity feeds. *Applied Energy* 136, 649–61.
- [22] Abdulbari HA, Ling FWM, Hassan Z & Thin HJ (2018), Experimental investigations on biopolymer in enhancing the liquid flow in microchannel. *Advanced Polymer Technology*.
- [23] Abdulbari HA & Ming FLW (2015), Drag Reduction Properties of Nanofluids in Microchannels. *The Journal of Engineering Research* 12, 60.
- [24] Abdulbari HA, Wang Ming FL & Mahmood WK. (2017), Insoluble additives for enhancing a blood-like liquid flow in microchannels. *Journal of Hydrodynamic* 29, 144–53.
- [25] Ling FWM & Abdulbari HA (2017), Enhancing the Flow in Microchannel using Natural Polymeric Additives. *Indian Journal of Science Technology* 10, 1–5.
- [26] Ling FWM, Mahmood WK & Abdulbari HA (2017), Rapid Prototyping of Microfluidics Devices using Xurography Method. *MATEC Web Conference* 111, 01009.
- [27] Ling FWM & Abdulbari HA (2017), Drag reduction by natural polymeric additives in PMDS microchannel: Effect of types of additives. *MATEC Web Conference* 111, 01001.
- [28] Abdulbari HA & Ling FWM (2017) Hibiscus mucilage for enhancing the flow in blood-stream-like microchannel system. *Chemical Engineering Communication* 204, 1282–98.
- [29] Porada S, Sales BB, M Hamelers HV, Biesheuvel PM, Sales B & Hamelers H (2012), Water Desalination with Wires. *The Journal of Physical Chemistry* 3, 1613–8.
- [30] Suss ME, Baumann TF, Bourcier WL, Spadaccini CM, Rose KA, Santiago JG (2012), Capacitive desalination with flow-through electrodes. *Energy & Environmental Science* 5, 9511.
- [31] Schlumpberger S, Lu NB, Suss ME & Bazant MZ (2015), Scalable and Continuous Water Deionization by Shock Electrodialysis. *Environmental Science & Technology Letters* 2, 367–72.
- [32] Suss ME, Porada S, Sun X, Biesheuvel PM, Yoon J & Presser V. (2015), Water desalination via capacitive deionization: what is it and what can we expect from it? *Energy & Environmental Science* 8, 2296–319.
- [33] Grygolowicz-Pawlak E, Sohail M, Pawlak M, Neel B, Shvarev A & de Marco R (2012), Coulometric Sodium Chloride Removal System with Nafion Membrane for Seawater Sample Treatment. *Analytical Chemistry* 84, 6158–65.
- [34] Roelofs SH, van den Berg A & Odijk M (2015), Microfluidic desalination techniques and their potential applications. *Lab on Chip* 15, 3428–38.
- [35] Pakielka Z, Ludwichowska K, Ferenc J & Kulczyk M (2014), Mechanical properties and electrical conductivity of Al 6101 and 6201 alloys processed by hydro-extrusion. *Materials Science and Engineering*.
- [36] Shahid M, McDonagh A, Kim JH & Shon HK (2015), Magnetised titanium dioxide (TiO) for water purification: preparation, characterisation and application. *Desalination Water Treatment* 54, 979–1002.
- [37] Boyer RR (1996), An overview on the use of titanium in the aerospace industry. *Material Science and Engineering* 213, 103–14.
- [38] Litvin DA & Smith DE (1971), Titanium for Marine Application. *Naval Engineers Journal* 83, 37–44.
- [39] Oryshchenko AS, Gorynin IV, Leonov VP, Kudryavtsev AS, Mikhailov VI & Chudakov EV (2015), Marine titanium alloys: Present and future. *Inorganic Materials: Applied Research* 6, 571–9.
- [40] Dupuis J, Chenon M, Faure S, Razan F & Gloriant T (2013), Mechanical properties and corrosion resistance of some titanium alloys in marine environment. *MATEC Web Conference* 7, 1009.
- [41] Sotto A, Boromand A, Balta S, Darvishmanash S, Kim J & Van der Bruggen B (2011), Nanofiltration membranes enhanced with TiO nanoparticles: a comprehensive study. *Desalination Water Treatment* 34, 179–83.