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Desalting of RO Waste and Drainage Water by Direct Contact Membrane Distillation

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ABSTRACT

Direct contact membrane distillation is an effective method for production of fresh water from saline water. In this study two samples were used as feed solutions; the first one was RO waste from Al-Hilla Coca-Cola Factory (TDS= 2382 mg/l) and the other was Haji Ali drainage water (TDS= 4127 mg/l). Polytetrafluoroethylene (PTFE) hydrophobic membrane supported with polypropylene (PP) was used as flat sheet form with plate and frame cell. Results proved that membrane distillation is an effective technique to produce fresh water with high quality from brine with low salinity content. With membrane area of 8x8 cm², the volume of treated water decreased from 34.97 ml at first half hour to 33.02 ml after 180 min of an experiment for first feed type and from 36.92 ml to 33.84 ml for the second feed type due to the fouling accumulated on the membrane surface. The temperature on both sides of the membrane surface and TDS of permeate was measured every 30 min of experiment time, also some tests were made on feed and permeate ions. Time of experiment was 180 min with the same operating conditions.

Key Words: direct contact membrane distillation, desalting of brine solution, RO waste water, PTFE Membrane.

تحلية مياه نفايات من منظومة التناضح العكسي ومياه مبزل بتقنية التقطير بالأغشية بالاتصال المباشر

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الخلاصة

تعتبر طريقة التقطير بواسطة الأغشية بالاتصال المباشر طريقة فعالة لانتاج المياه العذبة من المياه المالحة. في هذه الدراسة تم اخذ نموذجين الاول عبارة عن ماء فائض من منظومة تناضح عكسي لمصنع الكوكا كولا في الحلة (قيمة الاملاح الذائبة الكلية = 2382 ملغم/لتر) والآخر ماء من مبزل حجي علي (الاملاح الذائبة الكلية = 4127 ملغم/لتر) كلقيم للمنظومة. الغشاء الذي تم استخدامه عبارة عن PTFE وهو نوع من الأغشية الطاردة للماء مدعم بطبقة من البولي بروبيلين على شكل ورق مسطح موضوع داخل خلية من نوع اللوح والاطار. اثبتت النتائج ان عملية التقطير بواسطة الأغشية هي عملية فعالة لانتاج مياه عذبة ذات نوعية عالية من مياه قليلة الملوحة نسبيا. بمساحة غشاء مستخدم عبارة عن 8x8 سم²، وجد ان هنالك نقصان في حجم الماء المنتج من 34.97 مل الى 33.02 مل خلال زمن التجربة والبالغ 180 دقيقة بالنسبة للنموذج الاول، اما النموذج الثاني فان حجم الماء المنتج تناقص من 36.92 مل الى 33.84 مل بالنسبة للنموذج الثاني

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ويرجع السبب في ذلك الى تجمع النفايات على سطح الغشاء. تم قياس درجة الحرارة على جانبي سطح الغشاء وقياس نسبة الاملاح الذائبة الكلية للماء المنتج كل نصف ساعة من زمن التجربة، كذلك تم اجراء عدة فحوصات للماء المنتج والقيم. كان زمن التجربة 180 دقيقة مع مراعاة العمل بنفس الظروف التشغيلية.
الكلمات الرئيسية: التقطير بالاعشوية بالاتصال المباشر، تحلية المياه المالحة، فضلات منظومة التناضح العكسي، الغشاء من نوع PTFE.

1. INTRODUCTION

Increased risks of depletion and pollution of fresh water sources on the earth are found **Raut, and Kulkarni, 2012**. Desalting is defined as the process which isolated the salts from aqueous solution to get fresh water, **Shatat, and Riffat, 2013**. It was found from an early time as an active way to produce fresh water; the ancient seafarers used it on ships to get their needs of water, **Birkett, 1984**. Many process deal with desalting of brine solution as a thermal process (distillation), membrane process (RO, NF, etc.), and the combination of thermal and membrane process (MD, freezing, and solar dehumidification), **Boulianne, 2017**. Thermal and membrane processes need high energy (to get heating or pressure) which mean high cost, so it is necessary to look for other ways to decrease the cost. Membrane distillation focused on this side (decreasing cost) by using waste or solar energy, so many types of research were done to demonstrate its effectiveness. Desalting by membrane distillation (MD) is defined as a process where the hydrophobic membrane is used to separate fresh water from brine solution at a wide range of temperature and concentration; this process operation conditions can deal with a temperature less than boiling point degree and atmospheric pressure. The process is summarized by the existence of hydrophobic membrane between two sides where feed (hot) with direct contact with membrane for all types of MD and the other side of membrane either permeate (cold) or air. The difference in vapor pressure at both sides of the hydrophobic membrane caused by temperature difference is the driving force in MD mechanism, vapor molecules of fresh water moved across the membrane pores to the permeate side, **Drioli, et al., 2009**. In direct contact membrane distillation (DCMD), the feed (brine solution) and permeate (fresh water) are in direct contact with membrane sides and only vapor cross the membrane from feed side to permeate side where it condenses due to cold temperature of permeate side compared with feed side temperature, **Phattaranawik, et al., 2003** as shown in **Fig. 1**. The main advantages of using DCMD are using low energy, simple design, and high separation efficiency, while the disadvantages are low production according to other separation types, heat loss by conduction relatively high, and effect of temperature polarization, **Martinez, and Florido, 2001**.

The purpose of this experiment was to find the efficiency of using membrane distillation as a treatment method for the large quantity of wastewater produced from other freshwater production operation methods as RO, also for desalting the moderate saline sources like drainage sources.

2. EXPERIMENTAL WORK

2.1 Experiment System Contents

The experimental scale of a DCMD system was designed and installed in the laboratory as shown in the schematic diagram of **Fig. 2**. The membrane cell was plate and frame instilled in it flat sheet of hydrophobic PTFE membrane (Sterlitech Company-Kent-USA) supported by a woven (scrim) of polypropylene (PP) with an effective area of (0.0064 m²) with the following properties: total thickness (PP+PTFE) = 203 microns, void volume = 90%, pore size = 0.2



µm, and with operation temperature of 232°C. Three pumps were used to provide permeate, feed, and cooling water. Heater is submerged in feed tank with the controller to maintain the feed required temperature. Submerged coil provides the cooling temperature to permeate tank with the controller to keep the required temperature for condensing the penetrated vapor at permeate tank, which has an overflow to measure produced water in permeate tank. Four thermometers were used to measure inlet temperature and the temperature inside membrane model in both flow lines. Four pressure gauges used to measure pressure at inlet and outlet for both lines. Feed and permeate were controlled by valves (for measured and adjust flow) with two rotameter to measure flow.

2.2 Operating Conditions

New membrane was installed for every feed and citric acid solution with 10 wt. % was used for washing feed line to remove the accumulated salts (scaling and fouling) online. 37,47, and 57°C is feed temperatures (T_F), while 17°C used as permeate temperature (T_P), feed flow rate (Q_F) was 50 l/h and permeate flow rate (Q_P) was 40 l/h, with co-current flow, the pressure in feed side less than 0.05 bar and in permeate side almost atmospheric pressure. Operation conditions were kept to be constant at all experiment time.

2.3 Feed and Permeate Solutions Properties

Two feed solutions were used in experiments. One of them was RO waste water from Al-Hilla Coca-Cola Factory with total dissolved salts (TDS) of 2382 mg/l and other properties are shown in **Table 1**. The other feed solution was collected from Haji-Ali drainage water (Babil/Al-Hilla/Abou-Gharaq) with total dissolved salts of 4127 mg/l and other properties are shown in **Table 2**. Permeate solution was distilled water prepared from one step laboratory distillation device; the distilled water has the specific properties listed in **Table 3**.

2.4 The Measured Parameters

Results were measured every 30min, with 180 min as operating time. The parameters were flux, TDS for permeate side, inlet and outlet pressure for permeate and feed line, the temperature at feed and permeate tank, temperature before enters the model for permeate and feed, and the temperature inside the model for both sides of the membrane. After every experiment, some tests were made for residual feed and accumulated permeate by using many devices as shown in **Table 4** and titration methods, also through all the time of experiment random TDS test done by portable and bench type TDS meter for permeate to be sure there is no penetration for feed solution through the membrane.

3. RESULTS AND DISCUSSION

3.1 Effect of Feed Temperature

MD is a non-isothermal process so the effect of temperature is an important parameter to be studied, in both feed types with increasing feed temperature flux increase as shown in **Fig. 3**; for Haji Ali Drainage feed with increasing feed temperature 10°C (from 37°C to 47°C) with constant permeate temperature the flux increasing slightly from 10.35 LMH to 10.72 LMH. This increase continues when the temperature increases more 10°C (from 47°C to 57°C) to reach 11.32 LMH. For second feed (RO waste), the flux increase from 10.61 LMH to 10.92 LMH for the first 10°C increasing in temperature, also with increasing the temperature from 47°C to 57°C, the flux increases from 10.92 LMH to 11.76 LMH. This increase is due to the



increased vapor pressure in feed side which in turn increases the vapor of the flux that crosses through the membrane to permeate side. This increase also depends on the increase of diffusion of the penetrated molecules through the membrane, which is caused by the increase in thermal motion of membrane polymer chain, and the increase in temperature that makes permeate molecules more active and easy to diffuse according to the Eyring theory of diffusion and this is in agreement with the results of previous studies, **Hwang, et al., 2011, He, et al., 2011, Gryta, et al., 1997, and Adnan, et al., 2012**. The highest flux is when feed temperature (T_F) was 57°C , this behavior applies to the two types of feed used in experiments.

3.2 Effect of Time

Experiments time were 180 min, with calculating results every 30 min. The flux begins decreasing slightly with an increase in time. For the first feed of Haji Ali Drainage, the flux decreases from 11.54 LMH at the first 30 min to 10.58 LMH after 180 min, while for second feed (RO waste) the flux decreases from 10.93 LMH to 10.39 LMH for the same time. This decrease is due to the accumulation of fouling on membrane surface as shown in **Fig. 4**. Fouling and scaling lead to clogging in membrane pore, also they cause damage or wetting for the membrane, fouling caused by deposition of salts particles which they depend on the size of solids, this behavior is discussed by, **Kullab, 2011**; as in the effect of feed temperature; this is for both types of feed. The results agree with results studied by **Martínez, 2004**. When fouling is formed, this leads to decreasing vapor pressure which is caused by the increase in temperature polarization, **Martínez, and Vazquez, 1999**.

3.3 Effect of Concentration

The concentrations of feeds are different, so a comparison can be done to find the effect of concentration on flux. With increasing in feed concentration flux decrease as shown in **Fig. 5** which represent incorporation of **Fig. 4** (A&B). The flux decreases for feed with higher TDS (Haji Ali Drainage, TDS = 4127 mg/l) 10.93 LMH than the flux of RO waste feed (TDS = 2382 mg/l) which equals to 11.54 LMH at 30 min experiment time, this decrease exists in all experiment time. This increase is due to the fouling and scaling of membrane surface which reduces the vapor pressure on the feed side. The decrease in flux with increasing concentration is a result of decreasing in water activity coefficient, decrease in mass transfer coefficient, and a decrease in heat transfer coefficient, also increasing in feed concentration lead to decreasing in vapor pressure, flow, and temperature polarization (increased). This is due to the effect on feed activity and viscosity. This agrees with, **Alkudhiri, et al., 2012, Curico, and Drioli, 2005, Khayet, 2011**.

3.4 Permeate TDS Value

To examine process quality, permeate TDS was measured every 3 min. Results showing that desalting by DCMD is an effective process with an efficiency reaching 99.9% of salts removing.

3.5 Temperature in Both Sides of Membrane

Temperature was measured in both sides of membrane for both feed types to take view about temperature polarization; **Fig. 6** shows the temperature inside model for both feed types.



From the figure, as time increases the membrane will be wetted which increases the heat transfer through the membrane and this, in turn, will lead to decrease in flux due to a decrease in feed surface tension and also because wetting the membrane pore which must be mostly dry to facilitating the passage only for vapor. This was in agreement with, **Shirazi, et al., 2014**.

4. CONCLUSIONS

- 1- DCMD is an effective process for the desalting of an aqueous solution with low energy consumption.
- 2- The process has high efficiency with flux quality reaching 99.9% of the expulsion of salts.
- 3- With increasing the temperature, flux increases for all concentrations.
- 4- With increasing in time, flux begins decrease according to the formation of scaling and fouling.
- 5- With the increase in concentration, flux decrease due to accumulated salts on the membrane surface.

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NOMENCLATURE

DCMD= direct contact membrane distillation

LMH= liter per square meter per hour

MD = membrane distillation

NF = nano filtration

PP = polypropylene

PTFE = polytetrafluoroethylene

Q_F = feed flow rate, l/h

Q_P = permeate flow rate, l/h

RO = reverse osmosis

TDS = total dissolved solids, mg/l

T_F = feed temperature, °C

T_P = permeate temperature, °C

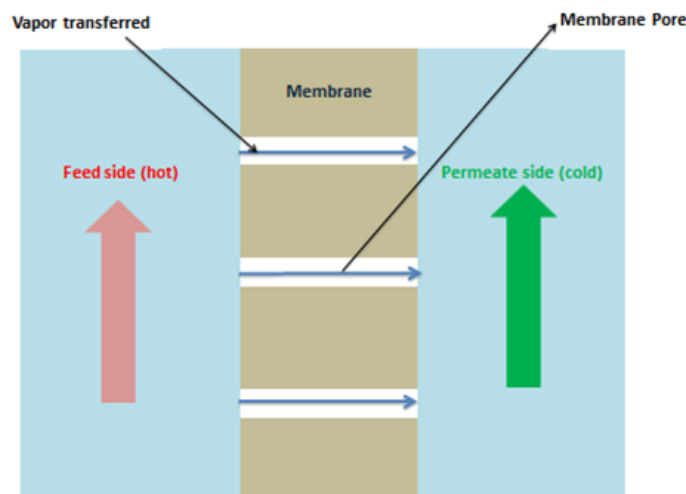


Figure 1. DCMD mechanism.

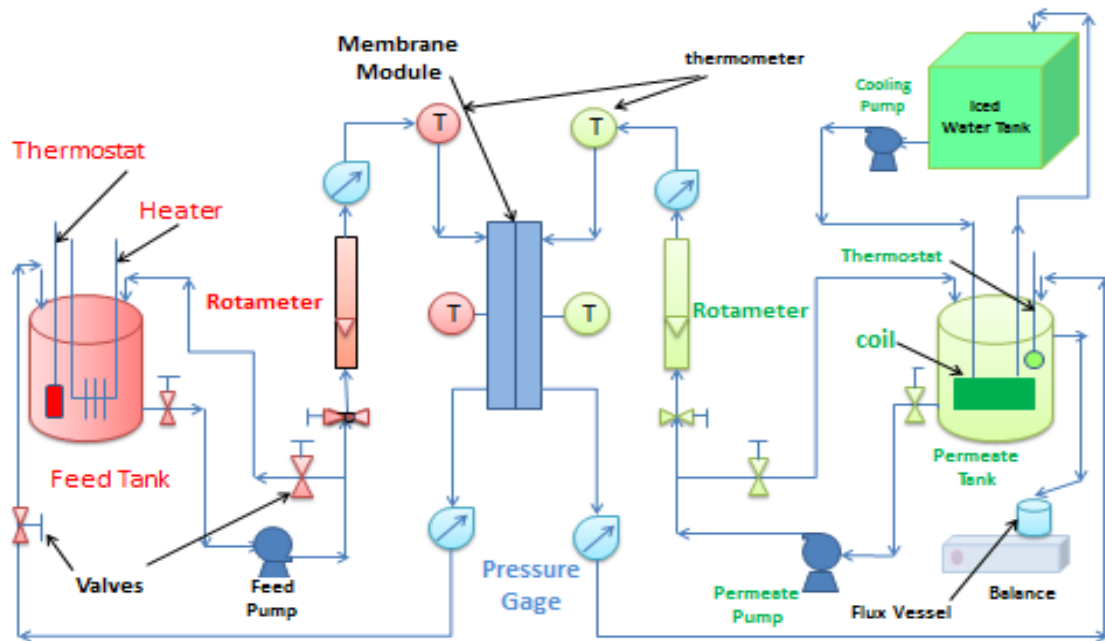
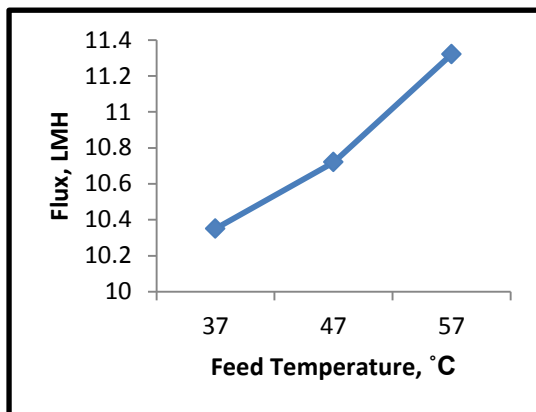
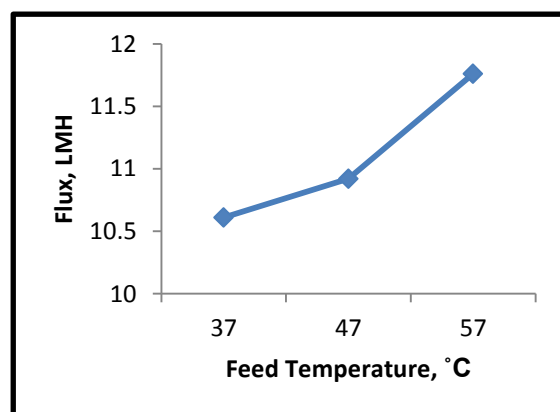


Figure 2. Schematic diagram for laboratory DCMD.

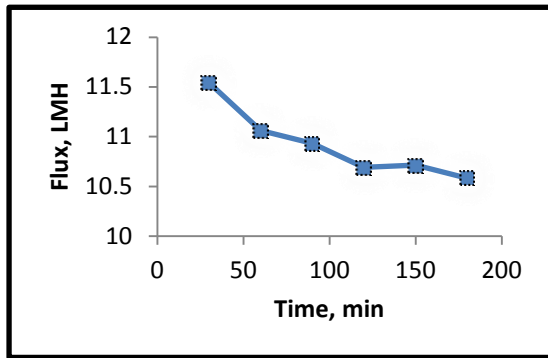


(A) Haji-Ali drainage feed

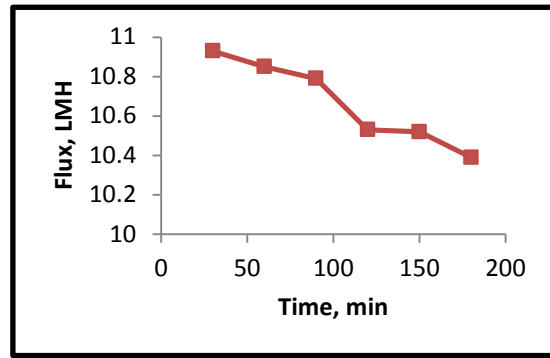


(B) Al-Hilla Coca-Cola Factory RO waste

Figure 3. Effect of Feed Temp. On Average Flux Amount ($Q_F=50$ l/h, $Q_P=40$ l/h, $T_P=17^\circ\text{C}$).

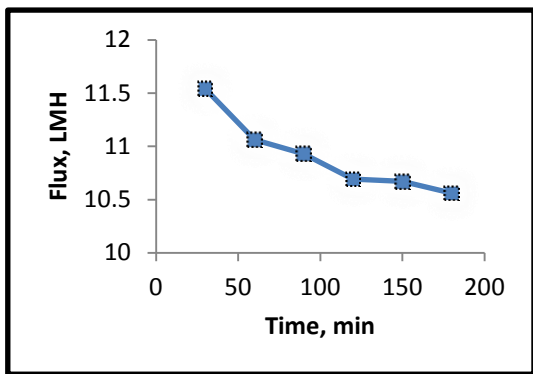


(A) Haji-Ali drainage feed

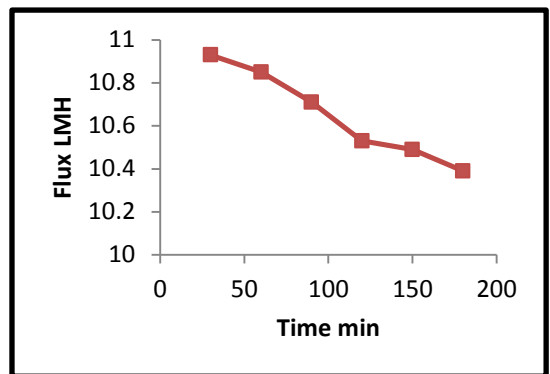


(B) Al-Hilla Coca-Cola Factory RO waste

Figure 4. Effect of time on flux amount ($Q_F=50$ l/h, $Q_P=40$ l/h, $T_F=47$, $T_P=17^\circ\text{C}$).

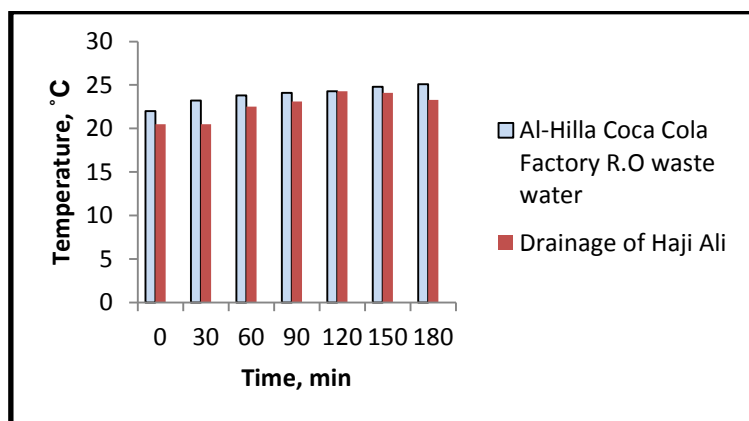


(A) Haji-Ali drainage feed

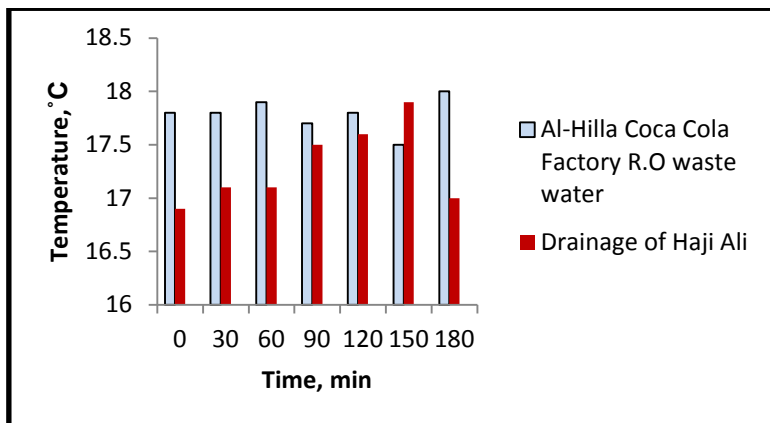


(B) Al-Hilla Coca-Cola Factory RO waste

Figure 5. Effect of feed concentration on flux amount ($Q_F=50$ l/h, $Q_C=40$ l/h, $T_F=47$, $T_P=17^\circ\text{C}$).



(A) Feed side



(B) Permeate Side

Figure 6. Effect of operating time with the temperature inside the module ($Q_F=50$ l/h, $Q_P=40$ l/h, $T_F=47^\circ\text{C}$, $T_P=17^\circ\text{C}$).

Table 1. Feed 1 properties.

| Parameter | Value mg/l |
|----------------|------------|
| TDS | 4127 |
| Sodium | 945 |
| Potassium | 25 |
| Total Hardness | 1942 |
| Chloride | 929 |
| Alkalinity | 400 |
| Magnesium | 65 |
| Calcium | 670 |

Table 2. Feed 2 properties

| Parameter | Value mg/l |
|----------------|------------|
| TDS | 4127 |
| Sodium | 945 |
| Potassium | 25 |
| Total Hardness | 1942 |
| Chloride | 929 |



| | |
|------------|-----|
| Alkalinity | 400 |
| Magnesium | 65 |
| Calcium | 670 |

Table 3. Distilled water properties.

| Parameter | Value mg/l |
|-----------------|------------|
| TDS | 2.1 |
| Sodium | Zero |
| Potassium | Zero |
| Total Hardness | 39.6 |
| Chloride | 1.03 |
| Alkalinity | 8 |
| Magnesium | 5.98 |
| Calcium | 6.03 |
| SO ₄ | 1.23 |
| pH | 6.5 |

Table 4. Devices used in tests.

| Device Name | Test | Properties |
|--------------------|---------------------------|-------------------------------------|
| Flame photometer | Na and K ions | Model BWB-XB/ United Kingdom origin |
| pH meter | pH | WTW- pH7110/ Germany |
| EC meter | TDS | WTW- Cond7110/ Germany |
| Portable TDS meter | TDS | Lab./China origin |
| Balance | Flux and analytical tests | Mettler Toledo/ Germany |