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MITIGATION OF SCALE PROBLEM
IN THE PUMPED DISI WATER TO AMMAN, JORDAN

Various methods are known to mitigate or prevent scale formation in pipes, rather by chemical addition, e.g., anti-scaling substances, or physically which includes ultrasonic or nanofiltration (NF). Nanofiltration membranes have a selectivity for the multivalent charged ions, so monovalent ions will pass the membrane partly and multivalent ions will be rejected completely. Chemical addition to prevent scale formation is based on justifying water parameters such as pH, alkalinity, and concentrations of ions that form the building units of scale crystal. In order to mitigate the scaling tendency in water pumped from the Disi aquifer to Amman city along its 345 km pipeline, different studies were conducted using simulated plumbing system. This part of the study is concerned with scale mitigation using nanofiltration and addition of chemicals. Nanofiltration was applied to reduce the hardness that causes scale deposition where it rejected around 70.5% of Ca^{2+}, 71.98% Mg^{2+}, 7.72% K^+, 29.0% Na^+, 66.63% Cl^-, 86.51% NO_3^-, 85.72% SO_4^{2-}, and 69.85% CO_2. Increasing the concentration of some ions such as Na^+, K^+ and Cl^- keeping the allowable limit gave good results for scale mitigation.

1. INTRODUCTION

Different methods are used to mitigate or prevent scale formation in pipes, rather chemical by addition of antiscaling substances, or physical which include ultrasonication or nanofiltration (NF). NF is a pressure-driven membrane process for liquid phase separations. This process has an excellent restriction on the passage of both organic and inorganic pollutants, including dissolved inorganic compounds, natural organic matter

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(NOM), bivalent or multivalent ions, and micropollutants [1]. NF process has advantages over other methods including low energy consumption and high flux rates [2].

The NF membranes lie between RO membranes (non-porous, where transport is governed by a solution-diffusion mechanism) and ultrafiltration (UF) membranes (porous, where separation is usually assumed to be due to size exclusion and, in some cases, charge effects). The separation mechanism of nanofiltration is based on both charge and size of particles [3, 4]. Nanofiltration membranes have a selectivity for the multivalent charged ions, so monovalent ions will pass the membrane partly and multivalent ions will be rejected completely [5].

Several studies have been conducted on the application of nanofiltration for water treatment. In 2002, nanofiltration was used for the treatment of ground water in Mainz, Germany, where ground water suffers from high hardness and high NOM content. Gorenflo et al. [6] used NF200B membrane characterized by high rejection of pesticides and organic matter as well as high rejection of calcium according to desirable purpose. A recent study carried by Al-Rawajfeh [7] proved that nanofiltration can be used as a deaeration step for the reduction of CO_2 gas that was released in MSF distillation.

Chemical addition to prevent scale formation is based on modifying water parameters, e.g., pH, alkalinity, and concentrations of ions that form the building units of scale crystal. Conventional methods used for prevention of scale deposition include: precipitation of salts using lime or soda ash, and addition of scale inhibitors. One of the simplest method is precipitation, where sodium carbonate is added causing the precipitation of calcium and magnesium ions carbonates. This step is followed by filtration before water is pumped through the distribution systems. Scale inhibitors are mainly phosphates and polyphosphates [8]. Diverse types of chemical additives are widely used in order to solve potable water problems. These chemicals are organic or inorganic substances. Generally, presence of organic and inorganic species in systems will modify solubility or stability of formed crystals. Some of these chemical additives are used to adjust pH such as sodium carbonate and sodium hydroxide,while other are used as corrosion and/or scale inhibitors, e.g., phosphates and polyphosphates. Phosphates perform numerous functions in drinking water treatment such as control of iron and manganese ions in water, removal/inhibition of scales, corrosion control, and stabilization of chlorine. In general, the function of most scale inhibitors is based upon suppressing the calcite crystal growth. Phosphate ions are adsorbed on active growth sites of calcite crystals, therefore, they inhibit and postpone the growth of calcite crystals. Phosphate based inhibitors are expensive and may lead to undesired effects such as eutrophication and algal blooms [9]. Water should be kept slightly scaling to ensure the existence of a protective film [10]. The formation or dissolution of calcium carbonate scale is dependent upon the concentrations of species existing in water, its temperature, and ionic strength [11].

The Disi water conveyance project was inaugurated in 2013 to improve living standards and to solve the water shortage problem due to high population growth in Amman city, the capital of Jordan. The Disi water is of high quality with total hardness of 140

mg $CaCO_3/dm^3$, alkalinity of 170 mg $CaCO_3/dm^3$, and pH of 8.03. Water is transferred through the 345 km steel pipes from the Disi aquifer in the south of Jordan to Amman city as shown in Fig. 1 [12].
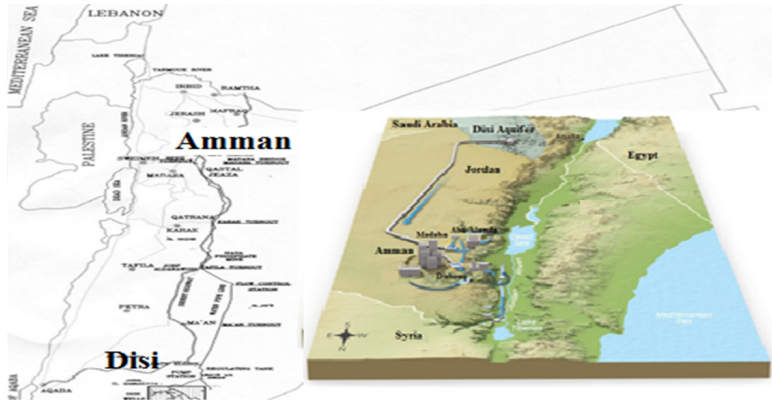

Fig. 1. Disi water conveyance project to transport water from the Disi aquifer to Amman city [12]

The Disi water is pumped to Amman city with different flow rates, one of them is under gravity. Figure 3 illustrates the direction of flow [12]. A previous study was undertaken to assess the quality of water from the Disi basin reaching consumers in Amman city. Moreover, the effect of mixing water from different sources was studied [13].

Water in plumbing systems suffers diverse problems, the most two important ones are: corrosion and scale formation. Some research was conducted in order to solve (reduce) these problems. Scale deposition is considered the major problem facing the pumped the Disi water in the network distribution system of Amman city. Scale film on the internal wall of the pipes in the plumbing systems is desirable due to its protective feature against corrosion. Nevertheless, over a period of time, such deposition will be accumulated and strengthen causing further problems concerned with flow rate efficiency and pipes clogging. To overcome the drop in water flow efficiency, more energy is required, as well as replacement of pipes may become necessary. Some studies showed that 1.0 mm thickness of scale can add 7.5% to energy costs, while 1.5 mm thick adds 15% of energy cost [14].

In this study, an experimental set up of 20.0 m pipes line was used to study the influence of flow rate on the Disi water chemistry, and how to reduce scale using nanofiltration and some chemical additives. Inorganic substances were used to mitigate calcite scale deposition on the inner walls of water pipes that comprise plumbing systems. The first part is based on changing the drinking water characteristics for some ions, thereby changing the solubility of some salts in the potable water. This treatment is based on inhibiting the growth of the calcite crystal, and on increasing the rate of dissolution of already suspended calcite precipitate [15]. For example, magnesium is commonly used in many studies in order to inhibit the nucleation, and crystal growth of

calcite [16]. Also, Mg^{2+} ions play an important role in the transformation process between polymorphs of calcite. Presence of Mg^{2+} ions reduces the transformation rate from the less stable morphism (aragonite) to the most stable morphism of $CaCO_3$ (calcite) [17, 18]. In the following sections, using inorganic substances as scale inhibitors will be considered.

2. MATERIALS AND METHODS

Based on standard methods of analysis [19], all collected samples were tested for physical and chemical parameters. pH was measured using a HI 254 pH/ORP meter. A conductivity meter (pH/ION/Cond 750) was used to measure electrical conductivity (*EC*). Total dissolved solids (*TDS*) were calculated based on the electrical conductivity. Total alkalinity (*TA*) was measured by the HCl titrimetric method, whereas for the total hardness (*TH*), calcium, and magnesium ion concentrations were measured by the EDTA titrimetric method. Dissolved carbon dioxide was measured by the NaOH titrimetric method. Argentometric titration was used for the chloride ion determination. Sulfate, nitrate, and phosphate ion concentrations were assessed using a UV-1800 Shimadzu spectrophotometer. Sodium and potassium ions were determined using the flame emission photometric method (Jenway PFP7 flame photometer).
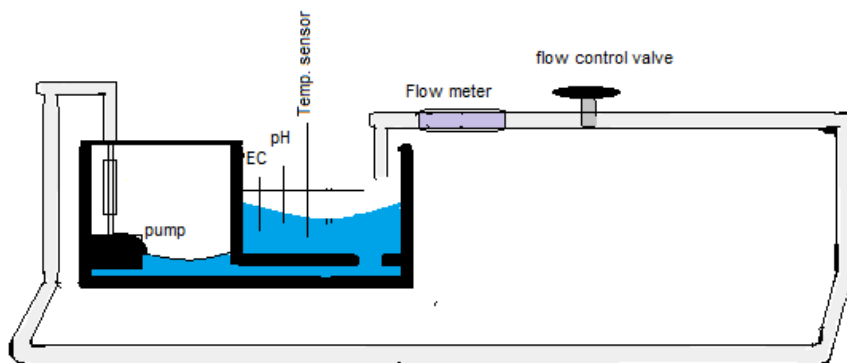

Fig. 2. Experimental setup for simulating the pumped Disi water

About 20.0 dm^3 of the Disi water was subjected to pumping through 20.0 m long pipes, 15 mm in diameter connected to a tank as shown in Fig. 2. Two methods of reducing scale have been investigated by simulation. The concentrations of substances that contributed in scale formation should be reduced in water. Ions that commonly participate in the deposition process are: calcium, magnesium, carbonate, sulfate, and phosphate ones. Table 1 shows the solubility products (K_{sp}) of the compounds that can be deposited. Phosphates of calcium and magnesium have the lowest K_{sp} values. Deposited phosphate salts are not taken into account due to their very low concentrations in pumped water. The most stable scale that can be formed is that of calcium carbonate (calcite)

with the K_{sp} value of 3.36×10^{-9}. So, this study is concerned with calcium carbonate scale.

Solubility products (K_{sp}) of common scale forming salts

Salt	CaCO$_3$ (calcite)	CaCO$_3$ (aragonite)	CaSO$_4$	Ca$_3$(PO$_4$)$_2$	MgCO$_3$	Mg$_3$(PO$_4$)$_2$
K_{sp} (25 °C)	3.36×10^{-9}	6.0×10^{-9}	4.93×10^{-5}	2.07×10^{-33}	6.82×10^{-6}	1.04×10^{-24}

Reduction of calcium and carbonate ion contents on water will reduce the deposited calcium carbonate scale as the following chemical equation shows:

Physical method – nanofiltration. Several physical methods can be used to mitigate scale formation. In this study nanofiltration is used in order to mitigate scale deposition in pipes of the plumbing systems. A nanofilter has been mounted in the simulated plumbing system used previously. A nannofltration system (Jordan Local Market) was connected to the pipes after the flow meter as illustrated in Fig. 3.
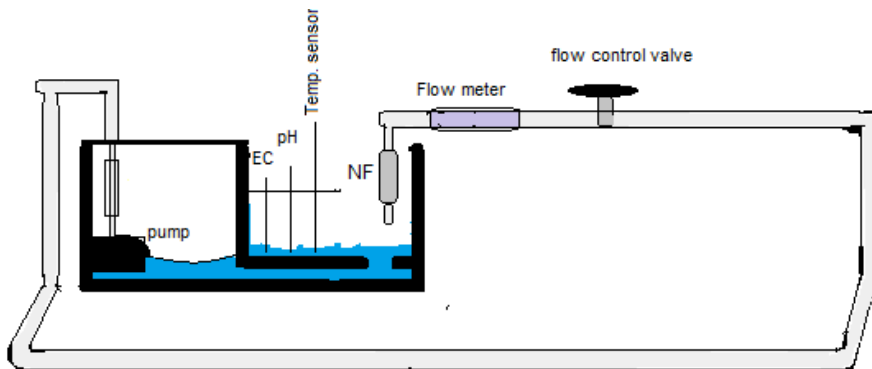

Fig. 3. Experimental setup for simulating the pumped Disi water using nanofiltration (NF)

Chemical additives. In this part, scale reduction has been conducted by the introduction of alkali chloride salts (NaCl and KCl) owing to the influence of these salts on the solute (Ca^{2+} and CO$_3^{2-}$) and water structure dynamics. Processes of calcite growth and dissolution can be affected by the solution composition. Both sodium and potassium ions are particularly used because they are essential to the human body. A 1.018 g of NaCl was dissolved in 10.0 dm^3 of the Disi water in the tank of the simulated plumbing system shown in Fig. 3. The added amount of NaCl increased the Disi water contents of sodium and chloride ions by 40.0 and 61.78 mg/dm^3, respectively, falling within acceptable limits of both JWDS and WHO standards. The modified water was then pumped for 16 h at the

flow rate of 35.0 dm^3/min. Temperature, pH, and conductivity have been measured directly every two hours.

Every 2 hours a sample has been collected to be analyzed for Ca^{2+}, Mg^{2+}, and dissolved CO_2 by the titrimetric method. Scale formation was monitored by estimation of the change in $[Ca^{2+}]$ using the EDTA titrimetric method. 10.0 dm^3 of the Disi water was pumped without any additives and under the same conditions for comparison purposes. The same experiment was repeated with the addition of 2.036 g NaCl in 10.0 dm^3 of the Disi water. This process increased the Disi water contents of sodium and chloride ions by 80.0 and 123.56 mg/dm^3, respectively. The concentration of both ions was maintained within acceptable ranges of both JWDS and WHO standards.

Two new experiments have been conducted under the same conditions using KCl instead of NaCl of two concentrations. In the former experiment, 0.191 g of KCl added to 10.0 dm^3 of water increased the Disi water contents of potassium and chloride by 10.0 and 9.10 mg/dm^3, respectively. In the latter experiment, 0.382 g of KCl was added to 10.0 dm^3 to increase the potassium and chloride ions contents by 20.0 and 18.20 mg/dm^3, respectively. Also concentrations of both ions fell within acceptable range of both JWDS and WHO standards

Finally, a mixture of NaCl and KCl has been used in the same manner. In this experiment, the contents of Na^+, K^+ and Cl^- were increased by 40.0, 20.0, and 79.98 mg/dm^3, respectively, by adding 1.018 g NaCl and 0.382 g KCl in 10.0 dm^3 of the Disi water.

3. RESULTS AND DISCUSSION

The Disi water was pumped through a nanofiltration system as shown in Fig. 3. The permeate water was then subjected to physicochemical analysis. The results are given in Table 2, the date for water samples before and after passing through the NF membrane can be found. From Table 2, it is clear that NF rejects about 70.5% of Ca^{2+}, 71.98% Mg^{2+}, 7.72% K^+, 29.0% Na^+, 66.63% Cl^-, 86.51% NO_3^-, 85.72% SO_4^{2-}, and 69.85% CO_2. Magnesium was rejected higher than calcium which is a consequence of the stronger hydration of the Mg^{2+} ion.

Measured parameters are shown in Fig. 4 for each sample collected and analyzed. There is an increase in both the temperature and pH values. The increase in temperature can be explained by the movement of water molecules and the friction with the internal walls of the pipes, while the increase in pH can be explained due to the fact that the solubility of gases decreases as the temperature increases. So the concentration of dissolved carbon dioxide (acidic) is expected to decrease during the pumping process and hence increase pH. A decrease in dissolved carbon dioxide concentration is also remarkable. Results showed a continuous decrease in $[Ca^{2+}]$ to a certain value followed by a fluctuation around it. This result shows the inhibition effect of the added Na^+ on the crystal growth of the scale.

Physicochemical properties of the Disi water
before and after filtration by nanofiltration

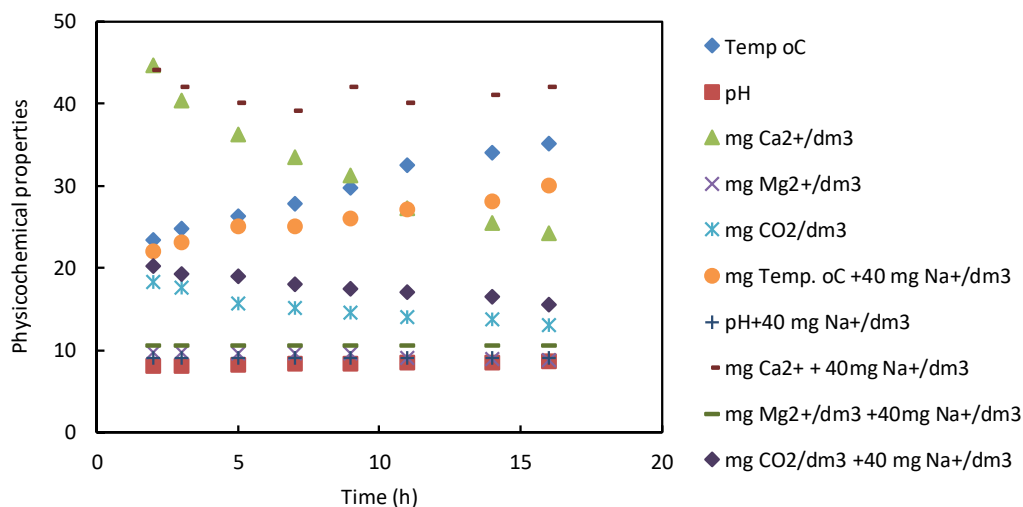| Property	Before filtration	After filtration
Temperature, °C	24.41	24.42
pH±0.001	8.030	7.630
TDS±0.1, mg/dm^3	210.6	157.9
TH, mg/dm^3	139.00±0.02	43.00±0.01
TA, mg/dm^3	170.00±0.03	161.53±0.02
[Ca^{2+}], mg/dm^3	40.00±0.02	11.80±0.04
[Mg^{2+}], mg/dm^3	9.60±0.04	2.69±0.01
[CO$_2$], mg/dm^3	17.60±0.01	5.31±0.00
[K$^+$], mg/dm^3	2.460	2.270
[Na$^+$], mg/dm^3	46.17	32.78
[Cl$^-$], mg/dm^3	39.05±0.03	13.02±0.05
[NO$_3^-$], mg/dm^3	13.20	1.780
[SO$_4^{2-}$], mg/dm^3	16.25	2.320

Fig. 4. Physicochemical parameters of the pumped Disi water
and Disi water with extra 40.0 mg Na$^+$/dm^3 as an additive

Based on Figure 5, similar trend was observed for the added amount of 80.0 mg Na$^+$/dm^3. Results showed a continuous decrease in [Ca^{2+}] to a certain value followed by progressive increase. This result improved the inhibition effect of the added Na$^+$ in addition to the dissolution action. The results for the Disi water without additions have been shown in each figure for comparison.

The set up used for simulation was cleaned with distilled water several times before each run. Sometimes diluted acid was needed to remove the deposited scale.
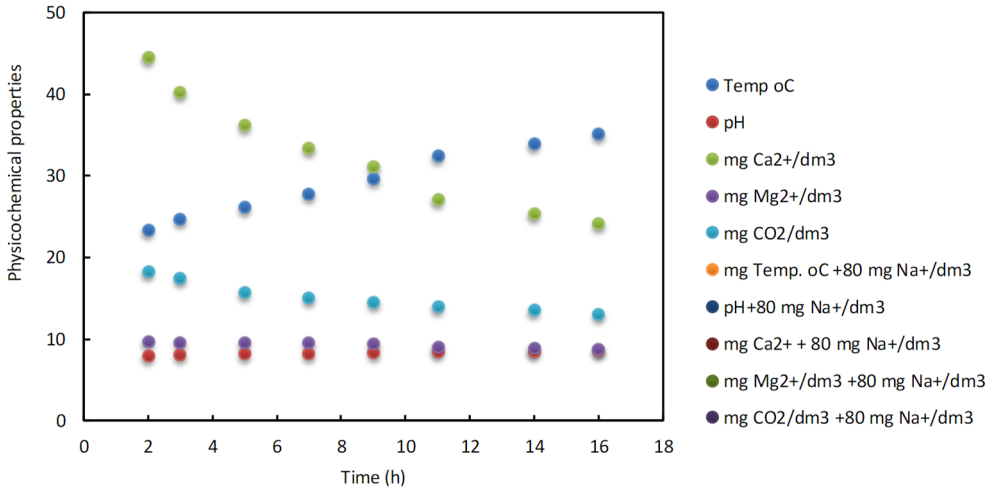
Fig. 5. Physicochemical parameters of the pumped Disi water
and Disi water with extra 80.0 mg Na^+/dm^3 as an additive

Figures 6 and 7 show all measured water parameters for the collected samples in the experiments with potassium ions used as an additive. In this case a similar trend was noticed for temperature, pH, and CO_2 but with a different degree of changeability. The results showed a continuous decrease in $[Ca^{2+}]$ for some time followed by slight increase in both experiments also. These results also indicate inhibition effect of the added K^+ in addition to the dissolution action.
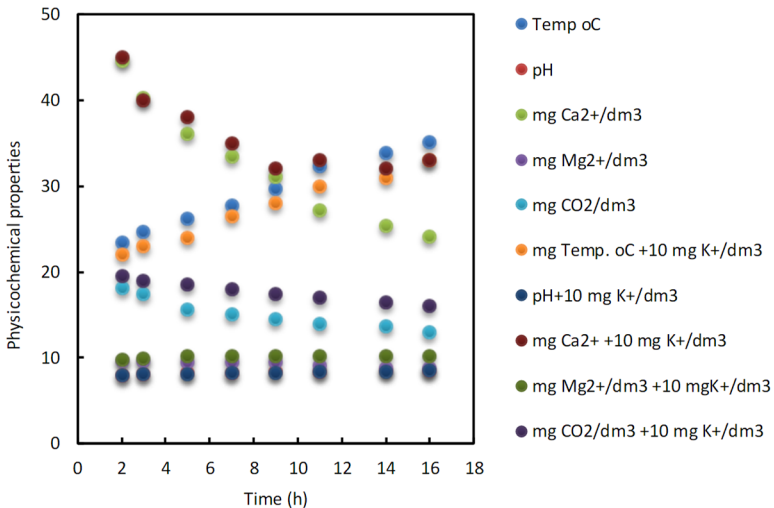

Fig. 6. Physicochemical parameters of the pumped Disi water
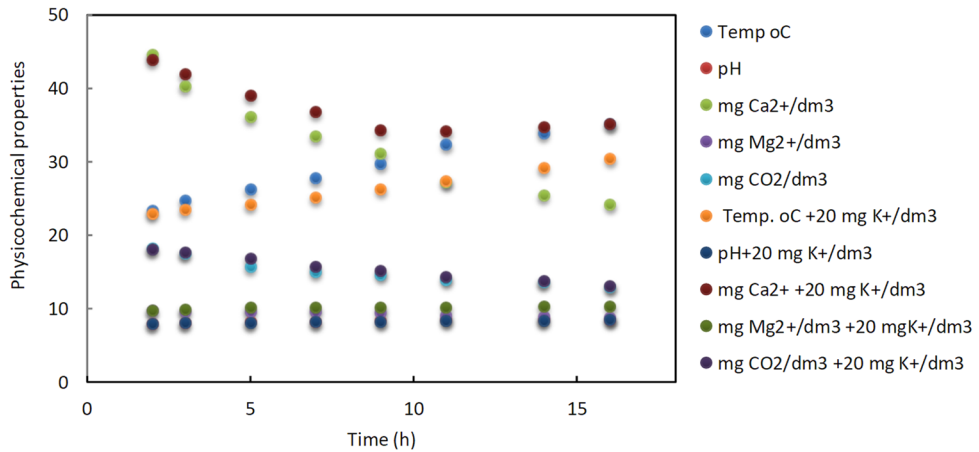and Disi water with extra 10.0 mg K^+/dm^3 as an additive

Fig. 7. Physicochemical parameters of the pumped the Disi water
and Disi water with extra 20.0 mg K$^+$/dm^3 as an additive

The results of experiments with a mixture of both Na$^+$ and K$^+$ ions are shown in Fig. 8. The results are similar as in the previous experiments with respect to trends observed, but differ in the degree of changes in the values of parameters. The temperature and pH increased, and the contents of carbon dioxide decreased upon time. It is also clear that the mixture of K$^+$ and Na$^+$ ions displayed effective inhibition and dissolution of scale better than K$^+$ and Na$^+$ ions alone in the concentration of 40 mg/dm^3.
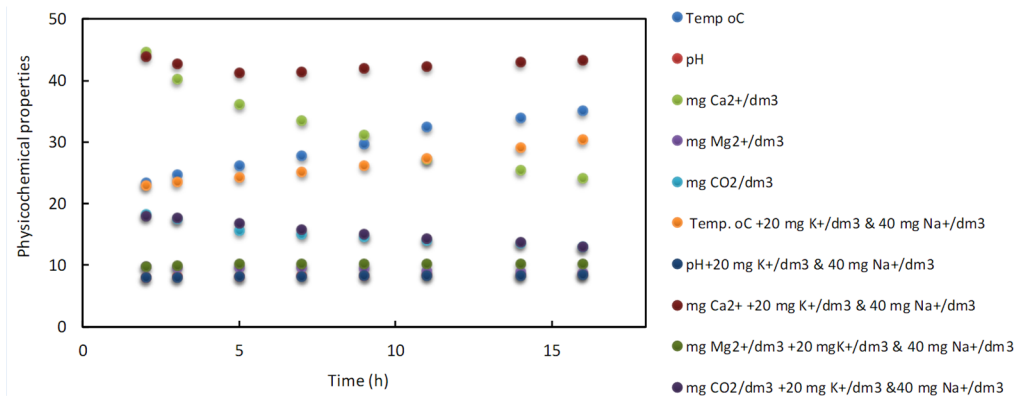

Fig. 8. Physicochemical parameters of the pumped Disi water
and Disi water with extra 20.0 mg K$^+$/dm^3, and 40.0 mg Na$^+$/dm^3 as additives

Scale formation or dissolution has been detected by following the concentration of Ca^{2+}. The concentration of calcium ion in mg/dm^3 was plotted versus time for all experiments in order to make comparison. Figure 9 shows the influence of used alkali chlorides on the concentration of calcium in the pumped Disi water.
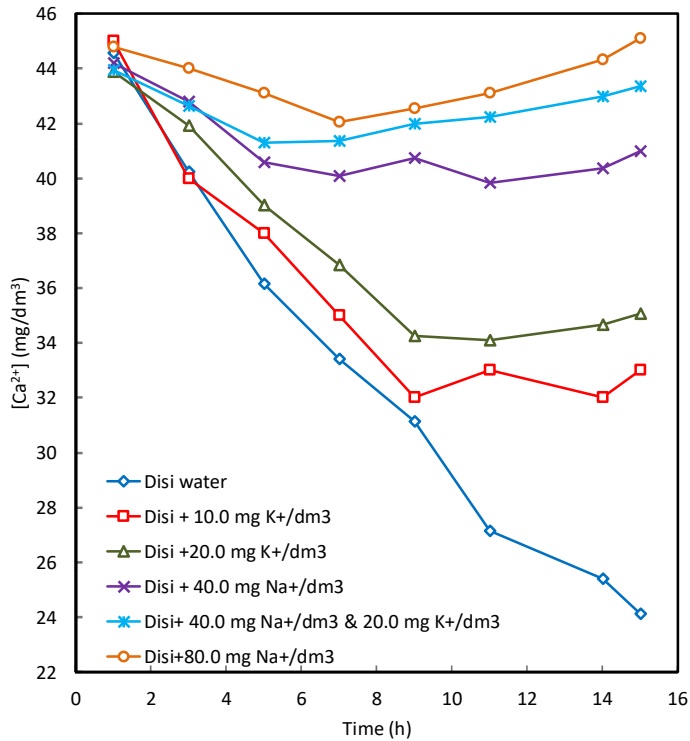
Fig. 9. Time dependences of the calcium ion concentration in the pumped Disi water
for various concentrations of alkali chloride salts (NaCl, KCl)

20.44 mg/dm^3. This decrease indicates the calcium carbonate scale formation during the pumping process. Accompanied with scale formation, there is noticeable increase in both temperature and pH . [Ca^{2+}] reduced progressively by the introduction of extra Na$^+$, and K$^+$ to a certain level, followed by slight increase to the end of experiment. With respect to 80.0 mg Na$^+$/dm^3, the increase in [Ca^{2+}] is recognizable, while for 10.0 mg K$^+$/dm^3 and 40.0 mg Na$^+$/dm^3 there is a fluctuation in [Ca^{2+}]. The fall down in [Ca^{2+}] was reduced in all cases of additives comparing to the Disi water only. The diversity of change in [Ca^{2+}] from salt to salt and from concentration to another is obvious too. The behavior of [Ca^{2+}] can be interpreted by two principal actions of the added salt. The first one is inhibition, and the second one is the dissolution.

Crystal growth and dissolution are governed by attachment and detachment of building units to/or from the crystal surface. Deposition and dissolution processes of salts including calcite in aqueous environment are strongly dependent on the rearrangement of water molecules around involved ions, and on the interaction between water molecules themselves. The rearrangement of water molecules around solute ions (Ca^{2+} and CO$_3^{2-}$) was considered to be the main kinetic barrier for both growth and dissolution

of crystals. Using simple ionic salts as additives will affect the behavior of crystals (dissolution or growth) through the ion hydration shell.

Type and amount of water-dissolved ions have strong influence on the formation, and growth of calcite crystals. The presence of Na^+ and K^+ ions in water lowers the electrostatic barriers of Ca^{2+} and CO_3^{2-}. The first interpretation of Ca^{2+} behavior launched from the perspective of crystal growth inhibition. K^+ and Na^+ ions stabilize the hydration shells of Ca^{2+} and CO_3^{2-} due to the electrostatic attraction between water dipole in hydration shells and counter ions existing in solution. Stabilization of ion hydration shells provides more separation between calcite building units (higher solubility). Better separation hampers the first step in crystal formation, which is the formation of ion pairs. So the presence of electrolytes obviously decreases the rate of calcite crystal growth. This interpretation illustrates the decrease in scale deposition, but it is noticed that, there is an increase in calcium ion concentration with time caused by dissolution of calcite already suspended in pumped water. For calcite to be dissociated, the affinity of water molecules to each other should be weakened, and the affinity of water molecules to calcite building ions should be strengthened. By strengthening the hydration of calcium and carbonate ions, the dissolution process becomes favored. Existence of electrolytes with high concentration leads to weakening the hydrogen bonds between water molecules which is also enhanced by the increase in temperature that accompanied the pumping process. This is expected to result in affinity of water molecules toward calcite crystal ions. Leading to increase in the dissolution rate of calcite crystals, thereby increase calcium ion concentration in water.

According to results in Figures 5–10, the presence of sodium and potassium ions with different concentration affects the change in $[Ca^{2+}]$ as the following order:

$$80 \text{ mg Na}^+/\text{dm}^3 > \text{mix } (40 \text{ mg Na}^+/\text{dm}^3 \text{ and } 20 \text{ mg K}^+/\text{dm}^3)$$

$$> 40 \text{ mg Na}^+/\text{dm}^3 > 20 \text{ mg K}^+/\text{dm}^3 > 10 \text{ mg K}^+/\text{dm}^3$$

It is obvious that Na^+ is more effective than K^+ to both crystal growth inhibition and dissolution. That can be explained due to the fact that sodium ions have a higher affinity to carbonate ion in calcite crystal than potassium ions. In other hands, Na^+ stabilized hydration shells of calcite building ions more than K^+ due to the greater affinity of Na^+ to water molecules than K^+ (i.e., Na^+ bonds to water molecules more strongly than to K^+). This could be attributed to the smaller Na^+ ionic radius (0.098 nm) than K^+ ionic radius (0.133 nm). Another observed result, is the consequences of concentration of the added ions. It was noticed as concentration of the added alkali chlorides increase, the efficiency of both inhibition and dissolution action increased. This is because of the increase in ionic strength leading to decrease in average crystal size which is governed by the total number of nuclei and the supersaturation of ions. The number of crystals that inhibited or crystals that dissolute also increased by increasing ions concentration.

A mixture of both alkali salts is used in order to maintain concentrations of ions in a suitable amount.

As a general reconnaissance, the presence of non-paired NaCl or/and KCl makes stabilization of water molecules in the hydration shell of calcite building units, leading to decrease the rate of crystal growth of calcite. Finally presence of chloride ions also increases water tendency for corrosion, but here in this study the concentration of chloride retained within the acceptable value.

Another way for the investigation of the mitigation in deposition tendency is what called stability indices that are used for the determination of water scale potential and corrosivity. Water is considered scale if it tends to deposit minerals such as calcium and magnesium insoluble salts. While water is considered corrosive if it tends to dissolve minerals [20]. Langelier saturation index (*LSI*) is the oldest, the most widely used index and is one of several parameters used for the predicting the corrosive properties of water [21]. It was puplished by Langelier in 1936 [21]. *LSI* is an equilibrium model derived from the theoretical concept of saturation, and used as a qualitative indicator of water tendency to precipitate or to dissolve calcium carbonate.

Zero value of *LSI* interpreted that water is at equilibrium. Positive value of *LSI* interpreted that calcium carbonate tends to deposit. While negative value of *LSI* indicates dissolution of calcium carbonate. Values greater than 1.5 or 1.7 represent warning of scale formation which will affect the chemistry of water and the flow of water through the pipes, while values less than –1.5 form indication of aggressive water which cause corrosion of pipes material and leads to the damage finally [22].

LSI was determined by the following relationship [21]:

$$LSI = pH - pH_s$$

where pH is the measured pH, pH_s is the pH at saturation in calcite or calcium carbonate and is defined as:

$$pH_s = (9.3 + A + B) - (C + D)$$

where:

$$A = \frac{\left(\log\left[TDS\right] - 1\right)}{10}, \qquad B = -13.12\log T + 34.55$$

$$C = \log\left[Ca^{2+} + CaCO_3\right] - 0.4, \qquad D = \log\left[alkalinity\ as\ CaCO_3\right]$$

LSI was calculated for all conducted experiments at the end of pumping process. Results were illustrated in Fig. 10. The decreases in *LSI* values with time is obviously clear for all additives.
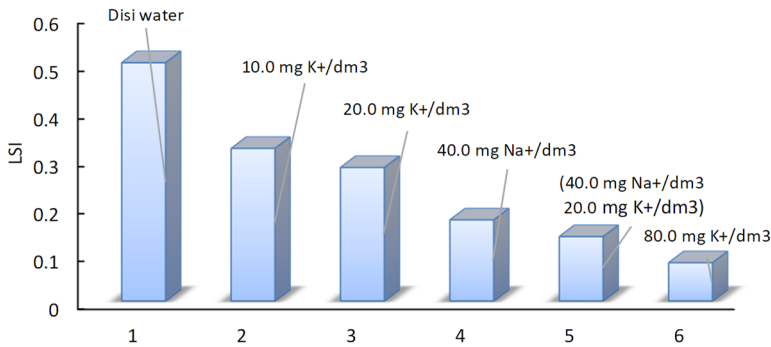
Fig. 10. The change in *LSI* values with different amount of alkali salts.

4. CONCLUSION

In order to mitigate scale formation in water pipelines, various methods have been examined in this study. Generally, nanofiltration is believed to reduce the concentration of ions that form the building units in scale crystals. Nanofiltration is considered to be an effective solution for scale formation problem in pipes of distribution systems. The results showed that adding alkali chlorides (NaCl and KCl) to the pumped the Disi water improved the simulation of scale mitigation. Various concentrations of both salts have been used and it was found that this significantly affected the rate of both the growth and the dissolution of calcium carbonate scale. As NaCl and KCl are the main components of the water system, there is no risk encountered from increasing their concentrations.

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