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# Investigating the Treatment of Oil and Gas Produced Water Using a Spray Dryer on a Bench Scale

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## Abstract

The current work investigates the performance of a single-stage, bench-scale system using a spray dryer to treat produced water. The produced water is generated in three large reservoirs of Ahvaz, Maroon, and Mansouri fields, which have different compositions but the same high total dissolved solids (TDS) and total organic carbon (TOC). The results of this study indicate that the newly developed bench scale rig is able to reduce the amount of TDS in the water produced in Ahvaz, Maroon, and Mansouri reservoirs to 98.78, 98.65, and 98.90, and TOC decreases the three types of the produced water to zero. Investigating the effect of independent parameters on the performance of this system using response surface methodology shows that the most effective parameters affecting the efficiency of the produced water treatment system are the entering carrier gas temperature (*TGIT*), the flow rate of the produced water (*QL*), the carrier gas flow rate entering the spray dryer (*QG*), and the atomizer pore size (*d*). Additionally, the optimal conditions are obtained as follows: *TGIT* = 113.7 °C, *QL* = 20.8 cc/min, *QG* = 59.9 $m^3$/hr., and *d* = 0.03 mm.

**Keywords:** Oil and Gas, Produced Water, Treatment, Spray Dryer

## 1. Introduction

A huge volume of produced water is obtained during the crude oil extraction, exploitation, and processing (Fakhru'l-Razi et al., 2009; Osipi et al., 2018). In the majority of the cases, produced water is either re-injected into deep wells (Minier-Matar et al., 2015) or discharged into reservoirs or seas (Khedr, 2011). Produced water is among the most important environment contaminants (Andrade et al., 2010). On the other hand, the oil-bearing regions mostly have very few water resources and are considered to be dry and arid regions in the world, so in recent years, reusing produced water has been a topic of interest.

In the oil fields in southwest Iran, produced water is either re-injected into the reservoir or discharged into ponds. The re-injection into the tank is a very expensive process due to the problems such as corrosion. In the case of discharge into ponds, the environmental consequences are severe. Therefore,

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the treatment of produced water to be reused can reduce the production costs and protect the environment.

Owing to the different types and amounts of impurity available in various oil reservoirs, the composition of produced water depends on the reservoir characteristics. Table 1 tabulates the substances usually found in oil reservoirs produced water along with some of their characteristics and their maximum and minimum level.

**Table 1**

Composition of oilfield produced water (Igunnu and Chen, 2012)

| Parameter | Minimum value | Maximum value | Heavy metal | Minimum value (mg/l) | Maximum value (mg/l) |
|---|---|---|---|---|---|
| **Density (kg/m$^3$)** | 1014 | 1140 | Calcium | 13 | 25800 |
| **Conductivity (mS/cm)** | 4200 | 58600 | Sodium | 132 | 97000 |
| **Surface tension (dyne/cm)** | 43 | 78 | Potassium | 24 | 4300 |
| **pH** | 4.3 | 10 | Magnesium | 8 | 6000 |
| **TOC (mg/l)** | 0 | 1500 | Iron | 0.1 | 100 |
| **TSS (mg/l)** | 1.2 | 1000 | Aluminum | 310 | 410 |
| **Total oil (IR; mg/l)** | 2 | 565 | Boron | 5 | 95 |
| **Volatile (BTX; mg/l)** | 0.39 | 35 | Barium | 1.3 | 650 |
| **Base/neutrals (mg/l)** | — | 140 | Cadmium | 0.005 | 0.2 |
| **Chloride (mg/l)** | 80 | 200000 | Copper | 0.02 | 1.5 |
| **Bicarbonate (mg/l)** | 77 | 3990 | Chromium | 0.02 | 1.1 |
| **Sulphate (mg/l)** | 2 | 1650 | Lithium | 3 | 50 |
| **Ammoniacal nitrogen (mg/l)** | 10 | 300 | Manganese | 0.004 | 175 |
| **Sulfite (mg/l)** | — | 10 | Lead | 0.002 | 8.8 |
| **Total polar (mg/l)** | 9.7 | 600 | Strontium | 0.02 | 1000 |
| **Higher acids (mg/l)** | 1 | 63 | Titanium | 0.01 | 0.7 |
| **Phenol (mg/l)** | 0.009 | 23 | Zinc | 0.01 | 35 |
| **Volatile fatty acids (mg/l)** | 2 | 4900 | Arsenic | 0.005 | 0.3 |
| | | | Mercury | 0.005 | 0.3 |
| | | | Silver | 0.001 | 0.15 |
| | | | Beryllium | 0.001 | 0.004 |

According to the diversity of the composition of produced water, various methods can be used to treat it (Igunnu and Chen, 2012; Jamei et al., 2017; Téllez et al., 2009). The results of a comprehensive study conducted on the water treatment methods in the oil and gas industry (Fakhru'l-Razi et al., 2009) show that choosing the best method depends on the properties of the feed (produced water), the amount of energy consumed, the cost-effectiveness of the method, the available space, the quality of the treated water, the operation continuity, and the quality of the byproducts.

The common methods for the treatment of produced water are significantly difficult and expensive. The work of Doran et al. on the performance of hybrid systems composed of a filter, an ion exchange module, and membrane on an industrial scale indicated that produced water used in the experiments have only 8000 ppm of TDS, which is needed for replacing the membranes in short operational intervals (Doran et al., 1998). A hybrid system consisting of a centrifuge, organoclay, microfiltration, ultrafiltration, and membranes has also been used by Burnett & Siddiqui (Sarker et al., 2006) to investigate the treatment of produced water in oil industries. They aimed to reduce the concentration of TDS and TOC respectively from 45000 and 200 ppm to standard agricultural water limits. This concentration is the highest amount of TDS and TOC reported among all the studies performed up to now, and it indicates the high performance of hybrid systems in treating highly contaminated produced water. The desalination and reuse of shale gas produced water with high-salinity were examined by Devin et al. (Shaffer et al., 2013) employing three methods of mechanical vapor compression (MVC), membrane distillation (MD), and forward osmosis (FO). Their results indicated that MD thermal method can be used for desalting produced water with a TDS concentration between 63000 and 500 ppm in regions where the energy costs are low, but the TOC reduction is not considered. It has also been demonstrated that, depending on the amount of the pollutants in water, the membrane-based methods are more economic and common than the other methods (Sanmartino et al., 2016); in other words, the membrane methods are incapable of water treatment at large concentrations of impurities due to the early fouling, and the thermal methods are preferred in such cases (Igunnu and Chen, 2012). Therefore, high water purification capability is the most important advantage of thermal methods (Hamed, 2004).

The most important difference between the membrane and thermal methods is the amount of consumed energy. Optimum thermal design method can lead to a considerable reduction in energy consumption (OPEX), a simultaneous decrease in capital expenditure (CAPEX), and using more affordable alloys for reducing the contact surface with corrosive media.

The byproduct of the common methods for the treatment of produced water is a highly polluted liquid (disposal). If a method is able to separate all or a fraction of the impurities in the treatment process of produced water, it can be expected that the environment will be less polluted. Research shows that spray dryers, operating based on the thermal rules, have not been used in any treatment processes of produced water (Fakhru'l-Razi et al., 2009).

In this study, the performance of the single-stage novel system was investigated, in which a spray dryer was used for the treatment of produced water. Additionally, the produced waters from three different crude oil desalination plants located in the southwest Iran, which all had a high concentration of TDS and TOC, have been used as the feed. Moreover, the effect of some parameters on the spray dryer was studied based on industrial and agricultural standards. Finally, the optimum condition was determined using a sensitivity analysis. The ability of the system to separate solid materials was also taken into account.

## 2. Materials and methods

### 2.1. Experimental setup

### a. Produced water treatment system

Figure 1 illustrates the bench scale setup consisting of a spray dryer, a cyclone, a condenser, a knock out drum (KOD), a fired heater, a blower, and a pump. In each experiment, the flow rate of the carrier gas is already adjusted after isolating the system using the blower remote regulator (Nanima motor SZ-09WM/1800W CLASS F), and it is monitored by an anemometer (Standard Anemometer ST-82) by considering the dimensions of the channel. Then, the temperature of the carrier gas passing through the heat exchanger is set for the fired heater, and the gas flow rate is set using a mass flow meter

(S420, CSi-tec, Germany). The produced water is controlled by the diaphragm pump pressure (C.C.K RO-900-EZ), and after adjusting and monitoring its flow by a variable area flow meter (OMEGA FLMH-1401AL), the produced water is converted into tiny drops with a diameter of 0.03 mm and distributed in the carrier gas inlet using an atomizer (Fixmee 3010 Brass Fog Mist Nozzle) in the spray dryer upper section. The spray dryer tower with a height of 420 cm and a diameter of 50 cm is made of galvanized plate with a thickness of 0.5 mm. The entire outer body is covered by glass wool thermal insulation with a thickness of 2.5 cm. The evaporated water and solids are removed from the channel located below the spray dryer. The fine grains of the dry matters dissolved by the cyclone are also separated from the carrier gas stream. The cyclone, which is made of the same material as the spry dryer, is designed by Wiliam F. Pentz (Pentz, n.d.) software. Afterward, the wet carrier gas is passed through the condenser to condense its moisture. The condensed liquid is separated from the carrier gas and accumulated in the KOD. Finally, the carrier gas flow is again connected to the blower by a five-inch channel.

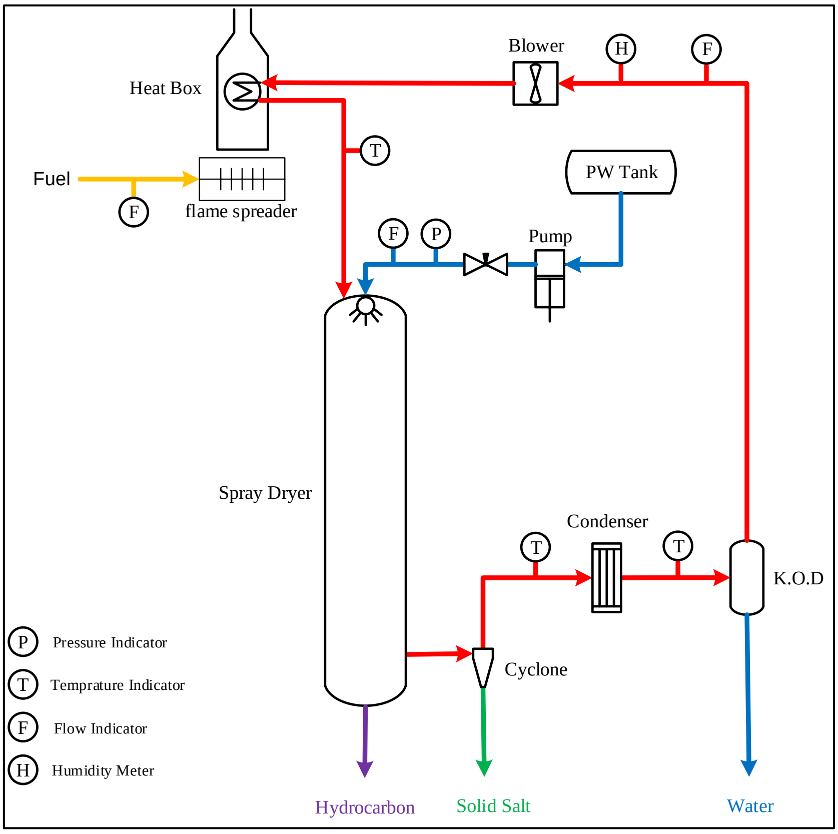



**Figure 1**

A schematic of the experimental setup.

The relationships governing spray dryers are extracted from the law of conservation of mass and energy and. Equation 1 expresses the heat transfer rate of the spray dryers (Czeslaw Strumillo, 1986):

$$Q_t = h_v . V_{ch} . \Delta\acute{T} \quad (1)$$

where, $h_v$, $V_{ch}$, and $\Delta\acute{T}$ are the volumetric heat transfer coefficient, volume of the drying chamber, and temperature gradient respectively.

The following relation also calculates the heat transfer coefficient in these types of towers (Czeslaw Strumillo, 1986):

$$h_v=2.13\times10^{-3}\frac{\lambda_g W_{m2}}{\rho_{m2} S_{ch}}\left(\frac{1}{\acute{d}_p}\right)^{1/6}\left(\frac{1}{u_{ge}+u_g}\right)^{0.8} \quad (2)$$

where, $\lambda_g$, $W_{m2}$, $\rho_{m2}$, $S_{ch}$, $\acute{d}_p$, $u_{ge}$, and $u_g$ are the heat conductivity coefficient of gas, dry product flow rate, output dry product density, cross-section area of the dryer chamber, dried particles diameter, final carrier gas velocity, and carrier gas velocity respectively.

Equation 3 calculates the height of the spray drier chamber as a function of its diameter:

$$H_{ch}=\frac{4V_{ch}}{\pi D_{ch}^2} \quad (3)$$

where, $V_{ch}$ and $D_{ch}$ are the volume and diameter of the spray dryer chamber respectively.

## b. Experimental methodology

Response surface methodology (RSM) was utilized to investigate the important effects of the parameters of the treatment process of the produced water influencing the volume of accumulated liquid in the spray tower and the volume of accumulated liquid in the KOD during the treatment. The seven independent variables were inlet carrier gas temperature to spray dryer tower, the flow rate of the produced water at the inlet, the circulation flow rate of the carrier gas, the temperature of the condenser outlet air, atomizers pore size, TDS, and TOC. The ranges of the process parameters were determined using preliminary experiments. A central composite rotatable design (CCRD) including 19 experiments arranged by 6 central points and 6 axial points to a $2^2$ full-factorial design was used. The experimental data were introduced to a linear model, and the regression coefficients were obtained for each answer. There were significant terms in the model found by the analysis of variance (ANOVA), and significance was suggested by the *F*-statistic calculated from the data. The competence of the model was confirmed by $R^2$, Adj-$R^2$, and Pre-$R^2$, where $R^2$ > 0.83 and (Adj-$R^2$ - Pre-$R^2$ < 0.2. After developing the model, other analyses, including the examination of the diagnostic plots and the calculation of the case statistics, were conducted to approve the assumptions made in ANOVA. The suitability of Design Expert software for optimizing systems, including spray dryers, has been demonstrated by reviewing previous literatures (Erbay et al., 2014). Design Expert Ver. 10 (2007) was utilized to provide response surfaces and optimize the treatment process. Simultaneously, the desirability function method was employed to optimize multiple responses.

In the present study, it is important to minimize the liquid accumulation at the bottom of the spray dryer and to maximize the accumulation of the condensed fluid in the KOD. Since an ideal system has the minimum energy consumption and maximum performance capacity, the importance of the temperature of the carrier gas entering the spray dryer and the flow rate of the produced water entering the spray dryer should be minimized and maximized respectively.

Table 2 lists the results of the experiments performed on the produced water of three different oil reservoirs, namely Ahvaz, Maroon, and Mansouri reservoirs, using the bench scale setup. It should be noted that the duration of each test was 20 minutes.

**Table 2**

Experimental data on the treatment of the produced water of Ahvaz, Maroon, and Mansouri reservoirs.

| Run | TGIT (°C) | QL (cc/min) | QG ($m^3$/hr.) | TGOC (°C) | d (mm) | TDS (ppm) | TOC (ppm) | LVAT (cc) | LVAK (cc) |
|---|---|---|---|---|---|---|---|---|---|
| **1** | 140 | 40 | 60 | 12 | 0.03 | 92415 | 115 | 0 | 800 |
| **2** | 140 | 45 | 60 | 12 | 0.03 | 92415 | 115 | 5 | 895 |
| **3** | 140 | 50 | 60 | 12 | 0.03 | 92415 | 115 | 1 | 999 |
| **4** | 140 | 55 | 60 | 12 | 0.03 | 92415 | 115 | 5 | 1095 |
| **5** | 140 | 60 | 60 | 12 | 0.03 | 92415 | 115 | 150 | 1050 |
| **6** | 140 | 40 | 60 | 12 | 0.06 | 92415 | 115 | 60 | 740 |
| **7** | 140 | 50 | 60 | 12 | 0.06 | 92415 | 115 | 100 | 900 |
| **8** | 140 | 50 | 40 | 12 | 0.03 | 92415 | 115 | 140 | 860 |
| **9** | 140 | 40 | 40 | 12 | 0.03 | 92415 | 115 | 20 | 780 |
| **10** | 140 | 60 | 40 | 12 | 0.03 | 92415 | 115 | 250 | 950 |
| **11** | 140 | 45 | 30 | 12 | 0.03 | 92415 | 115 | 80 | 820 |
| **12** | 140 | 45 | 50 | 12 | 0.03 | 92415 | 115 | 20 | 880 |
| **13** | 140 | 45 | 50 | 20 | 0.03 | 92415 | 115 | 70 | 830 |
| **14** | 140 | 45 | 50 | 30 | 0.03 | 92415 | 115 | 100 | 800 |
| **15** | 140 | 45 | 60 | 12 | 0.03 | 105452 | 250 | 5 | 895 |
| **16** | 130 | 45 | 60 | 12 | 0.03 | 105452 | 250 | 90 | 810 |
| **17** | 110 | 45 | 60 | 12 | 0.03 | 105452 | 250 | 190 | 710 |
| **18** | 130 | 45 | 60 | 12 | 0.03 | 100475 | 2000 | 40 | 860 |
| **19** | 110 | 45 | 60 | 12 | 0.03 | 100475 | 2000 | 170 | 730 |

## 2.2. Sampling and analysis

According to the flow rate of the produced water entering the spray dryer, the liquid accumulated in the bottom section of the spray dryer and KOD and the solid salt residues from the bottom of the cyclone were collected after each run. The volumes of the liquid samples were determined using a graduated cylinder, and the liquid samples were then poured in polyethylene terephthalate (PET) containers and analyzed in a laboratory. Moreover, the solid samples were weighed using a digital scale (KERN EMB 500-1SS05) and placed in plastic bags.

The following standard experiments were used to determine the composition of the feeds and products. The samples were injected to chromatography devices containing 20 mmol of methane sulfonic acid (for cations) or 20 mmol of potassium hydroxide (for anions) to analyze the cations and anions before being transferred to the analysis column (CS12A 4 × 250 mm for cations and AS19 4 × 250 mm for anions).

After passing the column, the charged ions were stopped by stopper membrane and monitored using a conduction coefficient detector. The system was calibrated based on industrial standards (Thermo Fisher Scientific). The acidic samples were blown into a nebulizer chamber and then sent to plasma for atoms/ions excitement. The elements were finally detected based on their diffusivity specifications using a spectrometer.

Organic carbons were determined by injecting an amount of samples into a combustion tube (720 °C) and being converted into $CO_2$. The amount of $CO_2$ was measured using an NDIR detector. Inorganic carbons were monitored by acidifying the samples and blowing air onto them. The amount of the $CO_2$ resulting from the inorganic carbons was also measured using an NDIR detector. The total organic material was the sum of the reported amount of the organic and inorganic carbons.

The conductivity coefficient and pH were evaluated using an Orion 3 Star device. Furthermore, TDS was calculated using the electrical conductivity coefficient. The hardness was also measured based on the titration method, and turbidity was monitored using a turbidimeter (HANNA HI93703).

## 2.3. Experimental uncertainty

Since empirical measurements are usually prone to error, uncertainty analysis was performed to validate the accuracy of the experimental results, especially the empirical data. To this end, Holman relation (Holman, 2001) was used and predicted the empirical measurements and their overall uncertainties. It is worth noting that these analyses are based on instrumentation measurements. In the present study, temperature, air flow rate, gas flow rate, volume, and mass were monitored using the explained equipment. The overall uncertainty of these measurements was calculated separately. Table 3 tabulates the results of the uncertainty analysis of the experimental measurements.

**Table 3**

Uncertainties of the experimental measurements.

| Experimental measurements | Unit | Comment |
|---|---|---|
| **Temperature** | °C | ±0.21 |
| **Weight** | gr. | ±0.018 |
| **Velocity** | m/s | ±0.33 |
| **Time** | s | ±5 |
| **Mass flow** | l/min | ±0.012 |
| **Volume** | ml | ±7 |

# 3. Result and discussion

## 3.1. Experiments with pure water (preliminary experiments)

Preliminary tests using pure water were conducted in order to find the operating range and monitor the thermal performance of the produced water treatment system. The experiments were performed in three categories. Each test was also repeated three times, and the average result of each type of the experiment was regarded as the action criteria.

The temperature of the carrier gas and the flow rate of the pure water entering the spray dryer were kept constant at 120 °C and 60 cc/min respectively in the first category of the experiments, and their corresponding results are shown in Figure 2; however, the flow rate of the carrier gas entering the spray dryer varied in each experiment in the range of 40-80 $m^3$/hr. This category included five experiments, each of which lasted for 35 minutes. The results of this experiment show that the flow rate of air (the carrier gas) should be greater than 58 $m^3$/min to avoid the liquid accumulation in the bottom of the spray dryer. It seems that a decrease in the flow rate of the carrier gas ($u_g$) leads to an increase in $h_v$ in Equation 2, but, at the same time, reduces $\Delta T$ more significantly; $V_{ch}$ also increases in Equation 1. Thus, the required height ($H_{ch}$) for the spray dryer increases in Equation 3. As a result, the droplet moves away from the upper part of the spray to the lower part, does not dry completely, and is trapped down the dryer, which, over time, causes liquid accumulation in the bottom of the spray dryer.

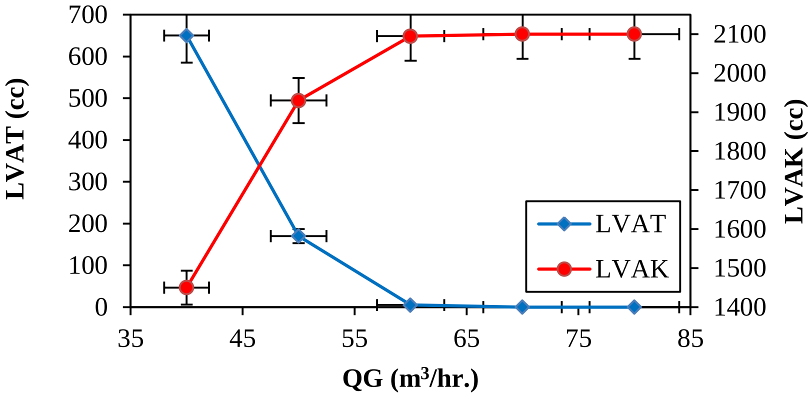



**Figure 2**

The effect of the flow rate of the carrier gas on the accumulation of water at the bottom of the spray dryer and the KOD; conditions: pure water, *TGIT* = 120 °C, *QL* = 60 cc/min, and *TGOC* = 12 °C.

The flow rates of the carrier gas and the pure water entering the spray dryer were kept constant at 60 $m^3$/hr. and 60 cc/min respectively in the second category of the experiments, and the corresponding results are displayed in Figure 3; however, the temperature of the carrier gas entering the spray dryer varied in each experiment in the range of 50-100 °C. This category included five experiments, each of which lasted for 35 minutes. The results indicate that the temperature of the carrier gas entering the spray dryer should not be lower than 59 °C in order to prevent water accumulation in bottom of the spray dryer. Also, a reduction in ΔT results in an increase in $V_{ch}$ (Equation 1). Therefore, the required height ($H_{ch}$) of the spray dryer in Equation 3 is increased. The low height of the spray dryer causes the droplets not to be completely dried before leaving the spray dryer and to remain in the spray dryer.

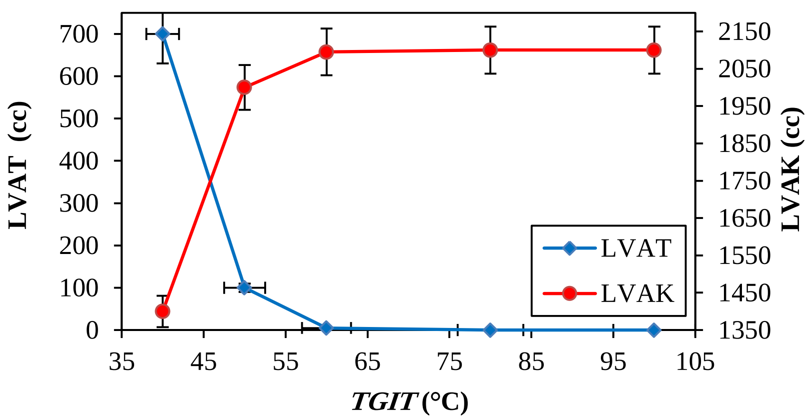



**Figure 3**

The effect of the temperature of the carrier gas on the spray dryer on the accumulation of water in the bottom of the spray dryer and the KOD; conditions: pure water, *QG* = 60 $m^3$/hr., and *QL* = 60 cc/min.

The flow rate and the temperature of the carrier gas entering the spray dryer were kept constant at 60 $m^3$/hr. and 120 °C respectively in the third category of the experiments, and the corresponding results are delineated in Figure 4; however, the flow rate of pure water entering the spray dryer in each experiment varied from 40 to 90 cc/min. This category included five experiments, and about 2000 cc of pure water was used in each experiment. The results demonstrate that, under the mentioned conditions, the pure water flow rate should be less than 60 cc/min so as to prevent water accumulation in the bottom of the spray dryer, which could be due to a decrease in ΔT caused by sudden evaporation. Therefore, $V_{ch}$ increases in Equation 1, which consequently raises the required height in Equation 3. As a result, droplets do not dry before exiting from the spray dryer and accumulate in the bottom of the spray dryer.

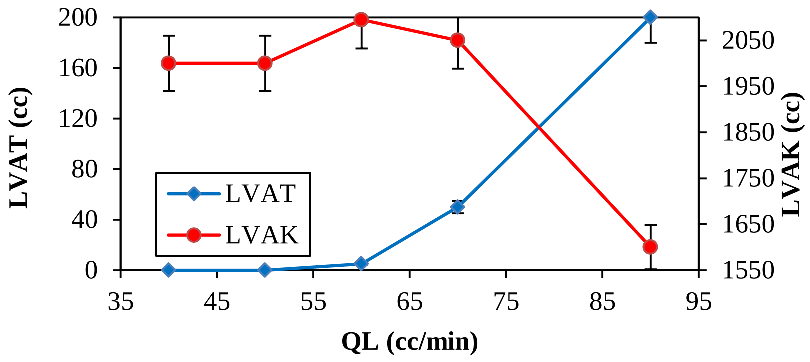



**Figure 4**

Effect of the flow rate of pure water entering the spray dryer on the accumulation of water at the bottom of the spray dryer and the KOD; conditions: pure water, *TGIT* = 120 °C, *TGOC* = 12 °C, *QG* = 60 $m^3$/hr.

## 3.2. Experiments with the produced water

The produced water used in the current study is the wastewater from the crude oil desalting units. Three types of the produced water from three oil reservoirs, namely Ahvaz, Maroon, and Mansouri, were obtained in the operation zone of National Iranian South Oil Company (NISOC) located in southwest Iran. The samples were collected and stored in 2.5-liter PET containers at ambient temperature (40 °C). Table 4 summarizes the composition of the produced water samples.

The results of the experiments with the produced water can be investigated from two points of view. In the first point of view, the system is analyzed for the quality of the product (treated water) and the amount of residual impurities. In the second viewpoint, the performance of the treatment system is evaluated.

**Table 4**

The composition of the produced water of Ahvaz, Marun, and Mansouri oil reservoirs.

| Parameter | Ahvaz field | Marun field | Mansouri field | Unit |
|---|---|---|---|---|
| **TDS** | 92415 | 105452 | 100475 | ppm |
| **Total hardness** | 28800 | 42300 | 45900 | ppm |
| **Temporary hardness** | 189 | 183 | 189 | ppm |
| **Permanent hardness** | 28611 | 42115 | 45711 | ppm |
| **Turbidity** | 559 | 387 | 246 | NTU |
| **pH** | 5.71 | 5.31 | 5.18 | - |
| **Calcium ($Ca^{+2}$)** | 9720 | 14400 | 15840 | ppm |
| **Magnesium ($Mg^{+2}$)** | 1094 | 3063 | 1531 | ppm |
| **Sodium ($Na^{+}$)** | 57927 | 66217 | 61036 | ppm |
| **Potassium ($K^{+}$)** | 590 | 699 | 586 | ppm |
| **Sulfate ($SO_4^{-2}$)** | 288 | 40656 | 37632 | ppm |
| **Chloride ($Cl^{-}$)** | 110050 | 107210 | 99400 | ppm |

| Parameter | Ahvaz field | Marun field | Mansouri field | Unit |
|---|---|---|---|---|
| **Total organic carbon (TOC)** | 115 | 250 | 2000 | ppm |

Tables 5 and 6 tabulate the analysis results of the obtained products, namely the treated water and separated solid salt. The existence of salt in the condensed water indicates that the salt microparticles exit from the spry dryer along with the carrier gas. However, the salt level (TDS < 2000) cannot prevent the treated water from being used for industrial and agricultural purposes. Moreover, the lack of carbon compounds (TOC = 0) in the materials discharged from cyclone and KOD reflects their entrapment in the spry dryer, which means that the carbon materials are completely removed in the water treatment process using the proposed method.

**Table 5**

The composition of the treated water (accumulated in KOD) obtained from the treatment of the produced water of Ahvaz, Maroon, and Mansouri reservoirs.

| Parameter | Ahvaz field | Marun field | Mansouri field | Unit |
|---|---|---|---|---|
| **TDS** | 1114 | 1421 | 1098 | ppm |
| **Total hardness** | 229 | 198 | 202 | ppm |
| **Temporary hardness** | 175 | 152 | 159 | ppm |
| **Permanent hardness** | 54 | 46 | 43 | ppm |
| **Turbidity** | 2.18 | 0.72 | 0.95 | NTU |
| **pH** | 7.1 | 7.2 | 7.1 | - |
| **Calcium ($Ca^{+2}$)** | 104 | 58 | 97 | ppm |
| **Magnesium ($Mg^{+2}$)** | 17 | 13 | 15 | ppm |
| **Sodium ($Na^{+}$)** | 268 | 434 | 294 | ppm |
| **Potassium ($K^{+}$)** | 103 | 110 | 103 | ppm |
| **Sulfate ($SO_4^{-2}$)** | 394 | 377 | 429 | ppm |
| **Chloride ($Cl^{-}$)** | 389 | 550 | 395 | ppm |
| **TOC** | 0 | 0 | 0 | ppm |

**Table 6**

The composition of salt (accumulated in cyclone) obtained from the treatment the produced water of Ahvaz, Maroon, and Mansouri reservoirs.

| **parameter** | Ahvaz field | Marun field | Mansouri field | **Unit** |
|---|---|---|---|---|
| **Sodium ($Na^{+}$)** | 295450 | 259787 | 302334 | ppm |
| **Calcium ($Ca^{+2}$)** | 47160 | 56880 | 60840 | ppm |
| **Magnesium ($Mg^{+2}$)** | 6564 | 6126 | 7220 | ppm |
| **Carbonate ($CO_3^{-2}$)** | 1320 | 1200 | 840 | ppm |
| **Sulfate ($SO_4^{-2}$)** | 16686.5 | 140503 | 16768.5 | ppm |
| **Chloride ($Cl^{-}$)** | 523711 | 500370 | 515673 | ppm |

Further, the results suggest that the TDS of the produced water of Ahvaz, Maroon, and Mansouri is reduced by 98.79, 98.65, and 98.90% respectively, which indicates that the studied system has a high capability to treat the produced water. It is also inferred that that the reduction in TDS is not affected by the TOC, which significantly differs among the three feeds. Therefore, the salts are independent of the carbon materials in the spray dryer. Table 7 also compares the performance of the current method with that of the methods used by other researchers in treating the produced water used in the present study.

**Table 7**

Comparison of the performance of the methods used for the treatment of the produced water.

| Method | Feed (ppm) | | Product (percentage of reduction) | | Researchers |
|---|---|---|---|---|---|
| | TDS | TOC | TDS | TOC | |
| **Pretreatment + adsorption/ion exchange + reverse osmosis** | 6000 | 120 | 97.58 | 98.33 | Doran et al. (Doran et al., 1998, 1997) |
| **Organoclay + centrifugation + reverse osmosis** | 45000 | 200 | 98.88 | 85.5 | Burnett et al. (Barrufet et al., 2005; Burnett, 2004; Sarker et al., 2006) |
| **Freeze-Thaw / Evaporation** | 12800 | - | 92.18 | - | Boysen et al. (Boysen and Boysen, 2008) |
| **Biologically active filtration + nanofiltration + reverse osmosis** | 18000 | 125 | 99.44 | 88.8 | Riley et al. (Riley et al., 2018) |
| **Pretreatment + vibratory shear enhanced processing + reverse osmosis** | 37500 | 1500 | 99.85 | 89 | Piemonte et al (Piemonte et al., 2015) |
| **Spray dryer** | 92415 | 115 | 98.78 | 100 | This work (Ahvaz) |
| **Spray dryer** | 105452 | 250 | 98.65 | 100 | This work (Marun) |
| **Spray dryer** | 100475 | 2000 | 98.90 | 100 | This work (Mansouri) |

In terms of the reduction in TDS, Table 7 confirms that the method proposed in the current research has better performance than the methods used by Doran et al. (Doran et al., 1998, 1997) and Boysen et al. (Boysen et al., 1999). Furthermore, the performance of the proposed method in reducing TDS is closely similar to that of the methods used by Burnett et al. (Barrufet et al., 2005; Burnett, 2004; Sarker et al., 2006), Riley et al. (Riley et al., 2018), and Piemonte et al. (Piemonte et al., 2015). It should also be noted that the amount of TDS in the feeds treated in the current study is much higher compared to the other works, and the applying membrane methods to treating such produced water faces fouling. In addition, the methods used by the other researchers are multistage processes which need pretreatments, while the method developed herein treats the produced water in a single stage. In the meantime, the proposed method, unlike the other methods, has completely removed the carbon materials (TOC), which is attributed to the nature of the method. In all the thermal methods, the polluted water is evaporated, so impurities are deposited. Thus, despite consuming a lot of energy, thermal methods are highly efficient.

Table 2 lists the results obtained by the experiments on the produced water taking account of the second viewpoint. This information is analyzed using Design Expert software version 10. The software also suggests a linear model which uses a regression test to predict the volume of the liquids accumulated in the bottom of the spray dryer and KOD. Equation 4 predicts the volume of the liquid at the bottom of the spray dryer:

$$\begin{aligned} LVAT = {} & intercept + C_{TGIT} \times TGIT + C_{QL} \times QL + C_{QG} \times QG + C_{TGOC} \times TGOC \\ & + C_d \times d + C_{TDS} \times TDS + C_{TOC} \times TOC \end{aligned} \quad (4)$$

And, Equation 5 presents the prediction of the remaining volume in K.O.D:

$$LVAK = intercept + C_{TGIT} \times TGIT + C_{QL} \times QL + C_{QG} \times QG + C_{TGOC} \times TGOC \\ + C_d \times d + C_{TDS} \times TDS + C_{TOC} \times TOC \tag{5}$$

Table 8 tabulates the regression equation constants of the obtained models and the important coefficients of all the significant process variables.

**Table 8**

Coefficients of the developed models.

| **Source** | LVAT (cc) | LVAK (cc) |
|---|---|---|
| | **Coefficient** | **Coefficient** |
| **Intercept** | +1142.35184 | -542.35184 |
| $\mathbf{C_{TGIT}}$ | -10.00000 | +10.00000 |
| $\mathbf{C_{QL}}$ | +8.46211 | +11.53789 |
| $\mathbf{C_{QG}}$ | -4.11351 | +4.11351 |
| $\mathbf{C_{TGOC}}$ | +3.83281 | -3.83281 |
| $\mathbf{C_d}$ | +2918.77368 | -2918.77368 |
| $\mathbf{C_{TDS}}$ | +4.20904E-004 | -4.20904E-004 |
| $\mathbf{C_{TOC}}$ | -0.021660 | +0.021660 |

The effect of the important process variables on each response was determined using analysis of variance as listed in Table 9. According to the results, the temperature of the carrier gas and the flow rate of the produced water entering the spray dryer with the respective *p*-values of 0.0014 and 0.0016 have the greatest impact on the volume of the liquids accumulated in the spray dryer and KOD, which is in agreement with Equations 1-3. On the other hand, parameters such as TOC and TDS with the respective *p*-values of 0.3219 and 0.08840 has the lowest influence on the volume of the liquids accumulated in the spray dryer and KOD.

**Table 9**

Analysis of variance of the linear evaluation for each response variable.

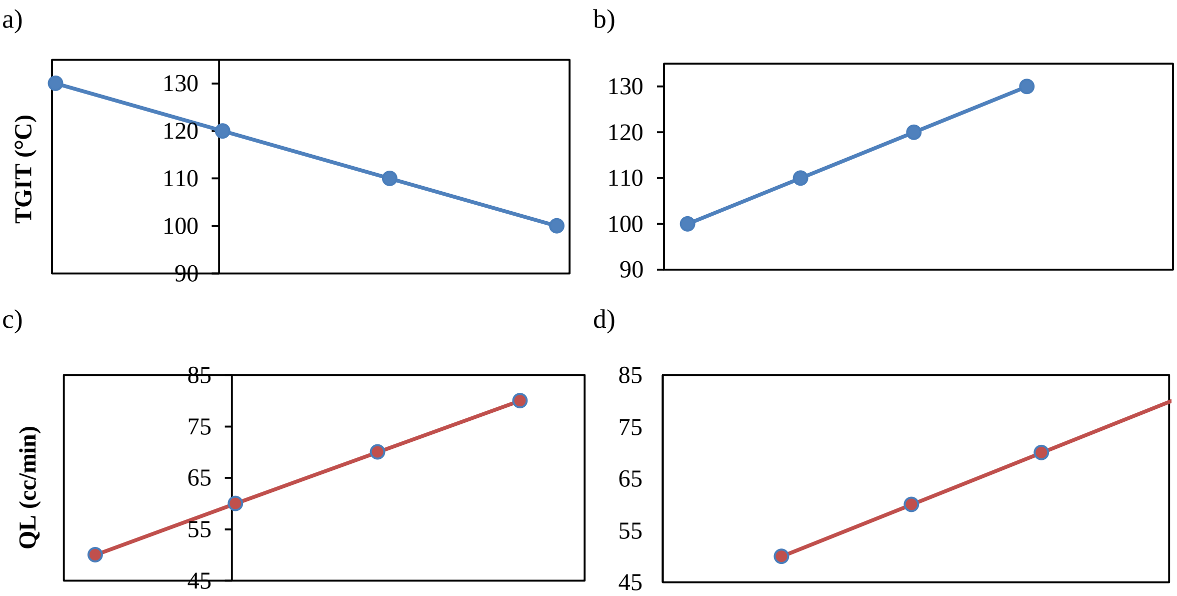
| **Source** | **Degree of freedom** | **LVAT (cc) Sum of squares** | **p-value** | **LVAK (cc) Sum of squares** | **p-value** |
|---|---|---|---|---|---|
| **Model** | 7 | 81246.08 | 0.0014 | 1.793E+005 | < 0.0001 |
| **TGIT** | 1 | 25000.00 | 0.0016 | 25000.00 | 0.0016 |
| **QL** | 1 | 38359.07 | 0.0003 | 71312.20 | < 0.0001 |
| **QG** | 1 | 19994.11 | 0.0035 | 19994.11 | 0.0035 |
| **TGOC** | 1 | 4674.71 | 0.1009 | 4674.71 | 0.1009 |
| ***d*** | 1 | 10610.21 | 0.0207 | 10610.21 | 0.0207 |
| **TDS** | 1 | 32.49 | 0.8840 | 32.49 | 0.8840 |
| **TOC** | 1 | 1568.39 | 0.3219 | 1568.39 | 0.3219 |
| **Residual** | 11 | 16039.61 | | 16039.61 | |
| **Total** | 18 | 97285.68 | | 1.954E+005 | |
| ***R*-Squared** | | 0.8351 | | 0.9179 | |

| | | |
|---|---|---|
| [a]**Adj *R*-Squared** | 0.7302 | 0.8657 |
| [b]**Pred *R*-Squared** | 0.5672 | 0.7845 |
| [c]**Adeq Precision** | 10.151 | 15.160 |
| [d]**Press** | 42101.10 | 42101.10 |

aAdjusted bPredicted cAdequate dPredicted Residual Error Sum of Squares

Figure 5 delineates the general trend in the variation of the response parameters as a function of the process variables using a fitted linear model. It is clear that the effect of the temperature of the carrier gas is more accentuated compared to the other independent variables. Hence, the temperature of the input carrier gas is considered as the most significant factor affecting the spray dryer process, which is completely evident in Equation 1 too. In fact, an increase in ΔT leads to a decline in $V_{ch}$, which consequently reduces $H_{ch}$ (Equations 2 and 3). The latter result has been confirmed by all the previous studies performed in spray towers (Bazaria and Kumar, 2016; Erbay, Z., Icier, 2009; Raimundo et al., 2017; Taylor et al., 2012). Other independent effective variables in the mentioned process sorted according to the degree of significance are QL, QG, and *d*. As demonstrated in Figure 5, the other independent variables also influence the amount of liquids accumulated at the bottom of the spray dryer and KOD. According to Equation 2, an increase in $W_{m2}$ enlarges $h_v$, which then reduces $H_{ch}$ (Equation 3). In this equation, $W_{m2}$ has an exponent equal to one, so it is also highly effective. However, the flow rate of the carrier gas entering the spray dryer has a less effect on the dependent variables of LVAK and LVAT since, in Equation 2, it is in the denominator with a power of 0.8. Moreover, as demonstrated in Equation 2, $d_p$ also negligibly affects $h_v$, and thus $H_{ch}$, because it is in the denominator with an exponent of 1/6.

Moreover, the data obtained from the experiments on the determination of the optimum conditions were analyzed using Design Expert software. It is concluded that the following conditions should be applied to the built-in water treatment system in order to minimize the amount of the liquid accumulated in the spray dryer, to maximize the volume of the liquid collected in the KOD, and to consume the minimum energy (minimum TGIT) for the maximum flow rate of the produced water entering the system (maximum QL): $TGIT$ = 113.7 °C, $QL$ = 20.8 cc/min, $QG$ = 59.9 m$^3$/hr., $TGOC$ = 12 °C, $d$ = 0.03 mm.



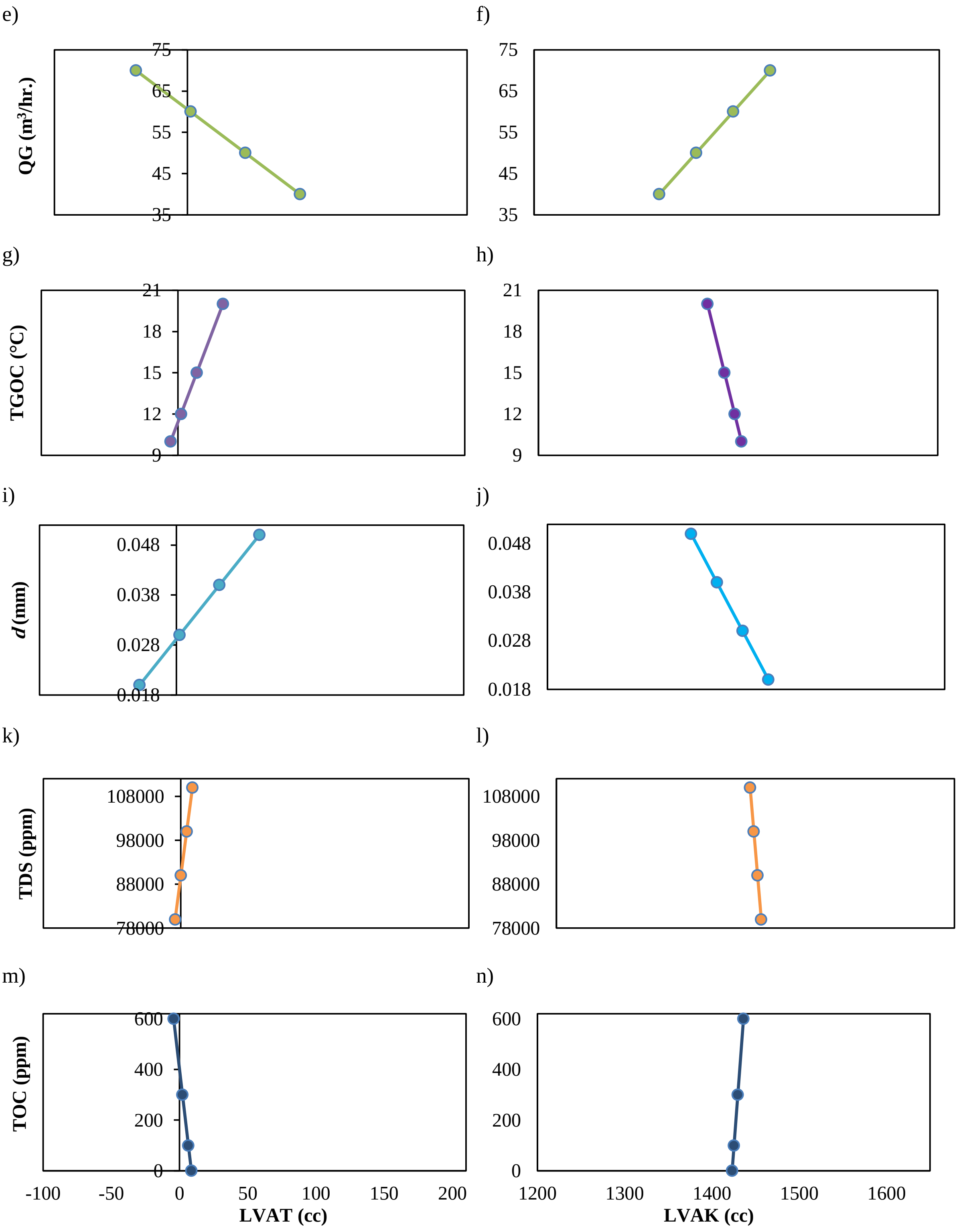



**Figure 5**

Variations in the liquid accumulated in the spray dryer and the liquid accumulated in the KOD versus the independent process variables using the developed models (Equations 6 and 7): (a and b) the temperature of the carrier gas entering the spray dryer; (c and d) the flow rate of the produced water entering the spray dryer; (e and f) the flow rate of the carrier gas entering the spray dryer; (g and h) the temperature of the carrier gas at the condenser outlet; (i and j) the hole size of the atomizer; (k and l) the concentration of TDS; and (m and n) the concentration of TOC.

## 4. Conclusions

The produced water treatment method developed in the current work exhibited a good performance; it reduced the TDS of the produced water of Ahvaz, Maroon, and Mansouri oil reservoirs by 98.78, 98.65, and 98.9% respectively and completely removed TOC from the produced water of all of the three reservoirs. The method was capable of eliminating the impurities of natural produced water in a single stage and producing clear water.

According to the results, the factors affecting the treatment process of the produced water in a system consisting of a spray dryer and a circulating carrier gas were the temperature of the carrier gas (air) entering the spray dryer, the flow rate of the carrier gas entering the spray dryer, the flow rate of the produced water entering the spray dryer, the atomizer pore size, the temperature of the carrier gas exiting from the condenser, and the concentration of the contaminants of the produced water (TDS and TOC). In this process, the temperature of the carrier gas entering the spray dryer (TGIT) was the most influential parameter affecting the treatment process of the produced water. It directly and highly impacted on both indicators of "liquid accumulated at the bottom of the spray dryer" and "liquid accumulated in the KOD". In the defined range of the experiments with natural produced water in this work, the optimum *TGIT* value was 113.7 °C. Also, the flow rate of the produced water entering the spray dryer (*QL*) was more effective on the studied indicators than the other factors. Moreover, the optimum *QL* value was equal to 20.8 cc/min. The third parameter impacting on the treatment process of the produced water was the flow rate of the carrier gas (*QG*), and the optimum *QG* value was calculated at 59.9 $m^3$/hr. Further, continuous salt extraction from produced water in a solid form was found possible in the present study.

The results indicated that the method used for the treatment of produced water can purify water and makes it suitable for industrial and agricultural use. One of the most important limitations to this method is the amount of the consumed energy, which requires precise investigations.

Considering the importance of energy consumption, in a future study, we will focus on the optimization of the energy consumption of the proposed method and compare it with other methods commonly used for the treatment of produced water such as membrane, distillation, etc.

## Nomenclature

| | |
|---|---|
| ANOVA | Analysis of variance |
| BTX | Benzene, toluene, and xylenes |
| $C$ | Constant |
| CAPEX | Capital expenditure |
| CCRD | Central composite rotatable design |
| $d$ | Diameter of nozzle hole |
| $d_p$ | Dried particles diameter |
| FO | Forward osmosis |
| $H_{ch}$ | Height of the drying chamber |
| $h_v$ | Volumetric heat transfer coefficient |
| IR | Infrared method |
| KOD | Knock out drum |
| *LVAK* | Volume of liquid accumulation in KOD |
| *LVAT* | Volume of liquid accumulation in tower |
| MD | Membrane distillation |

| | |
|---|---|
| MVC | Mechanical vapor compression |
| NDIR | Nondispersive infrared |
| NTU | Nephelometric turbidity unit |
| OPEX | Operating expense |
| PET | Polyethylene terephthalate |
| ppm | Part per million |
| $QG$ | Flow rate of the carrier gas |
| $QL$ | Flow rate of the produced water |
| RSM | Response surface methodology |
| $S_{ch}$ | Cross-section area of the dryer chamber |
| TDS | Total dissolved solids |
| TGIT | Temperature of the carrier gas entering the tower |
| TGOC | Temperature of the carrier gas at the condenser outlet |
| TOC | Total organic carbon |
| TSS | Total suspended solid |
| $u_g$ | Carrier gas velocity |
| $u_{ge}$ | Final carrier gas velocity |
| $V_{ch}$ | Volume of the drying chamber |
| $W_{m2}$ | Dry product flow rate |
| $\lambda_g$ | Heat conductivity coefficient of gas |
| $\rho_{m2}$ | Output dry product density |
| $\Delta T$ | Mean temperature |

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