

## Research Paper

# Contribute to the Removal of $Mg^{2+}$ From the Brine Produced by Reverse Osmosis Units Using $Na_3PO_4$

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**Abstract**— Desalination by reverse osmosis generates a stream of fresh water and approximately an equivalent volume of highly concentrated brine as waste. The waste brine is usually dumped back into the sea which increases the sea's salinity in a way that can affect marine life. In this context, the current study demonstrates the possibility of removing magnesium ions from the brine solution produced by reverse osmosis units, where the optimal conditions for precipitation of  $Mg^{+2}$  magnesium ions were studied using trisodium phosphate at a temperature of

$T = 20 \pm 5 \text{ C}^\circ$  and a pH = [8.5-9.4] corresponds to the output water. The research results indicate that when the ratio is:

$\frac{[PO_4^{3-}]}{[Mg^{2+}]} = 10$  we get a removal (95.9-97.3)% of magnesium ions, by adopting a mixing time of one hour, where the sedimentation

is maximum and corresponds to the balance of the two-phase system. The changes in calcium removal were studied in the presence of magnesium ions, where the removal yield decreased with the increase of magnesium ions in the solution, and it also led to the removal of the total hardness of the water by (92.6-97)% and the conversion of excessively hard water into soft water using trisodium phosphate, in addition to that the formed precipitate can be converted into fertilizer for plants by way to recycle it.

**Keywords**— reverse osmosis, brine, calcium ions, magnesium ions, removal, trisodium phosphate.

## 1. Introduction

There should be a lack of information in the previous paragraph. Desalination plants are changing in the Mediterranean Sea, causing impacts on the future marine environment [1], becoming at least 1.6 times more salinity than sea water, including the surrounding environment, including a comfortable, environmentally friendly and economically feasible environment for brines that Produced by mirrored osmotic volumes [2]. Generally, the resulting water is disposed of by draining into sanitary sewers, surface water discharges, deep well injection, and groundwater [3].

Most of the published literature discuss the potential impacts of desalination plants on the marine and terrestrial environment [4], [5], [6], [7], [8]. However, limited field research is available, particularly on the long-term effects on the marine environment. In brief, the potential impacts include: (i) Construction stage, which share its terrestrial effects with any other land development projects; (ii) Impact on the marine environment-high salinity brine discharge, chemical.

Reject brine from desalination plants contains various salts and ions, For the seawater reverse osmosis (SWRO) process, the concentration of these salts and ions in the reject brine (typically in the range of ~70,000 mg/L) is higher than their concentration in seawater (33,000–37,000 mg/L)[9]. Recent years have witnessed tremendous progress in the advancement of resource recovery technology from seawater and desalination brine[10].

## 2. Related Work

### I. water hardness:

The natural hardness of water comes from the dissolution of mineral geological formations containing calcium, magnesium, and silica, where this water can be oversaturated with one or more of the previous elements, which leads to the formation of (scaly sediments) in water distribution systems and heating technologies, especially in boilers. In general to hardness in the presence of calcium and magnesium compounds: bicarbonate ( $HCO_3^-$ ), carbonate ( $CO_3^{2-}$ ), sulfate ( $SO_4^{2-}$ ), chlorides ( $Cl^-$ ), and nitrate ( $NO_3^-$ ) in Water The silicates are derived from the dissolution of crystalline and

amorphous  $\text{SiO}_2$  and most of the clay minerals such as kaolinite, Illite, and Montmorillonite.

There are four main forms of scaly sediments: calcium carbonate ( $\text{CaCO}_3$ ), magnesium hydroxide ( $\text{Mg}(\text{OH})_2$ ), calcium sulfate ( $\text{CaSO}_4$ ), and silica ( $\text{SiO}_2$ ). Each deposit is formed from the previous deposits through sedimentation reactions at the normal temperature of surface and groundwater. With reference to the tendency of hard water to form scaly sediments with an increase in its temperature, and this is observed in heating technologies due to the inverse proportion between solubility and temperature, however, this inverse proportion quickly turns into a direct proportion when it comes to silica, as its solubility increases at degrees High temperature [11],[12].

In water, when  $\text{HCO}_3^-$  and  $\text{CO}_3^{2-}$  are the dominant anions compared to  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$ ,  $\text{CaCO}_3(\text{s})$  (calcite) will be the scaly precipitate formed in distribution systems Water and its heating technologies, but in the presence of a sufficient amount of  $\text{Mg}^{2+}$ , the scaly precipitate  $\text{Mg}(\text{OH})_2(\text{s})$  (Brucite) will form significantly compared to  $\text{MgCO}_3$  due to the disparity of solubility. If  $\text{SO}_4^{2-}$  are the dominant anions compared to  $\text{Cl}^-$ ,  $\text{HCO}_3^-$ ,  $\text{NO}_3^-$ ,  $\text{CO}_3^{2-}$ , then  $\text{CaSO}_4(\text{s})$  (Gypsum) is the squamous precipitate formed.

Silica under high pressure shows a clear tendency to form non-squamous deposits, but if calcium is present, the formed scaly precipitate can be calcium silicate. Squamous deposits, as shown in Figure(1), decrease economic feasibility and disadvantage technical performance [13].



Figure (1): Calcareous scaly deposits in water distribution systems

## II. Magnesium:

### Source:

Magnesium is found as a major component in the environment, including the earth's crust, ground and surface water, the marine environment, and fossil fuels (coal, oil, and natural gas), so that the elements appear in the water naturally, whose sources are usually associated with the basic natural processes that control the access of these elements to the water, which is chemical weathering for creeks and soil washing processes [14].

### Physical and chemical properties of magnesium:

Magnesium is an alkaline earth metal, which is the eighth most abundant in nature, and constitutes 3% of the earth's

crust. Due to its great effectiveness in entering into chemical reactions, it does not exist in a free form, but rather in the form of compounds, the most important of which are:

Magnesium Chloride  $\text{MgCl}_2$  21%

Carnallite  $\text{MgCl}_3 \cdot 6\text{H}_2\text{O}$  9%

Brucite  $\text{MgO} \cdot \text{H}_2\text{O}$  41%

Dolomite is  $\text{CaMg}(\text{CO}_3)_2$  14%

It is obtained in pure form by electrolysis of magnesium chloride (Dow's method) or by thermal reduction of magnesium oxide (Pidgeon method).

It is used in making some types of aircraft as it is the lightest of metals and is used to protect iron from rust and is used in various vital industries.

Symbol **Mg**

Atomic number **12**

The atomic weight is **24.3050**

### Magnesium toxicity:

Magnesium has great benefits for the human body, but several toxic symptoms emerged due to its excess than the required limit, as these symptoms emerged when it exceeded the permissible limit, the most prominent of which are:

Nervous system disorders.

Acute renal failure, especially for those who take magnesium-containing drugs

Weakness and paralysis of breathing.

An abnormality in the rhythm of the heartbeat [15].

Table (1): Permissible limits for magnesium in water according to WHO and Syrian standard specifications

water	Drinking water according to the World Health Organization WHO	Industrial water that flows into water bodies (sea water - rivers - water bodies) according to the Syrian standard specifications
The maximum allowable concentration	50 ppm	80ppm

## 3. Methodology

### I. Materials and devices used:

#### Materials:

- Magnesium Chloride(99.0-103.0%),  $\text{Mg}(\text{Cl})_2 \cdot 4\text{H}_2\text{O}$ , Calcium Nitrate(99.0-103.0%), Ammonium Chloride  $\text{NH}_4\text{Cl} \cdot 24\text{H}_2\text{O}$  (99%), Sodium Hydroxide (99%)  $\text{NaOH}$ , Disodium salt of  $\text{Na}_2\text{Y} \cdot 2\text{H}_2\text{O}$  ( $\text{Na}_2\text{EDTA}$ ) (99-100%), Aerochrome black indicator (200mg of it in 100g of  $\text{NaCl}$ ) and Miroxide indicator (200mg of it in 100g of  $\text{NaCl}$ ) are all manufactured by MERCK.
- Trisodium Phosphate  $\text{Na}_3\text{PO}_4 \cdot 12\text{H}_2\text{O}$  (97%), manufactured by TMMEDIA.
- Ethanol (99.7-100%).
- I used double distilled water from the ELGA system.

**The devices used:**

**Centrifuges** separate heterogeneous mixtures into their various components – liquids in liquids, solids in liquids, and liquids in gases, based on the different densities of the components. One of the most common uses is to separate red blood cells and other blood components from whole blood **Figure (2)**.

**pH Meter** is used to measure the acidity of the solution by comparing readings from a reference electrode and a sample electrode, the pH meters also measure temperature **Figure (3)**.

**Electrical conductivity** meter is an instrument for measuring electrical conductivity in solutions. This device is usually used to know the amount of salts in solutions and has practical uses in monitoring the amount of salts and impurities in water sources **Figure (4)**.



**Figure (2):**Antomed Centerfuge AM-8



**Figure (3):** The 744 pH Meter is manufactured by Metrohm..



**Figure (4):** Milwaukee Mi18 electrical conductor

## II. Research methods:

The conductivity, the amount of dissolved solid salts, the pH and the temperature in the output water generated by the reverse osmosis units in the General Company for Generating Baniyas were determined from the site for several consecutive months, then samples were taken to the laboratory for analysis.

A chemical and physical characterization of the output water was carried out.

The following ions were determined: calcium ion  $\text{Ca}^{2+}$  and magnesium ion  $\text{Mg}^{2+}$  using titration with a standard solution of  $\text{Na}_2\text{EDTA}$  at a concentration of (0.01M), then the optimal conditions for precipitation of magnesium ions using trisodium phosphate were studied, depending on the mixing time at the pH and temperature corresponding to the output water.

Finally, the optimum removal conditions were applied to the output water generated by reverse osmosis units.

### 1. Study of optimal conditions for the precipitation of magnesium ion $\text{Mg}^{2+}$ with trisodium phosphate $\text{Na}_3\text{PO}_4$ :

Solutions of the precipitating agent were prepared using  $\text{Na}_3\text{PO}_4$  trisodium phosphate:

$[\text{PO}_4^{3-}] = 0.05, 0.1, 0.5 \text{ M}$ , and magnesium ion solutions  $\text{Mg}^{2+}$  were prepared using magnesium chloride  $\text{Mg}(\text{Cl})_2$ :  $[\text{Mg}^{2+}] = 0.005, 0.01, 0.05, 0.1 \text{ M}$ , the required gradient in pH was obtained using solutions of acid perchlorine,  $\text{HClO}_4$ , and lithium hydroxide,  $\text{LiOH}$  [16], were measured with a 744 pH meter manufactured by Metrohm. (10 ml) of aqueous phases containing calcium ion  $\text{Ca}^{2+}$  was mixed with (1 ml) of aqueous phases containing different concentrations of trisodium phosphate  $\text{Na}_3\text{PO}_4$  in tubes of 15 ml made of polyethylene, then the samples were subjected to a centrifugation process at a speed of 3500 rpm. The concentration of calcium ions,  $\text{Ca}^{2+}$ , was determined using titration with the disodium salt of  $\text{Na}_2\text{EDTA}$  (Ethylene Diamine Tetra Acetic Acid) [17], and the changes in the yield of removing the cationic load from magnesium were studied

according to the mixing time at the pH of natural calcium ion solutions, which are pH=[8.5-9.4] and their temperature T=20±5C°.

**2. Study of optimal conditions for the precipitation of Ca<sup>2+</sup> in the presence of Mg<sup>2+</sup> ions:**

Solutions of magnesium ion Mg<sup>2+</sup> were prepared: [Mg<sup>2+</sup>]=0, 0.001, 0.005, 0.01, 0.05 M, each of which contains the calcium ion Ca<sup>2+</sup> with a concentration: [Ca<sup>2+</sup>]= 0.01M and the phosphate ion concentration  $\frac{[PO_4^{3-}]}{[Ca^{2+}]}=10$  and studied the

change in the removal yield of the calcium ion Ca<sup>2+</sup> according to the concentration of the magnesium ion Mg<sup>2+</sup> at pH = [8.5-9.4] and T = 20 ± 5C° for natural magnesium ion solutions Mg<sup>2+</sup> and time Mixing capacity 60 minutes.

**3. Application to output water samples generated from reverse osmosis units:**

The concentration of calcium ions was determined by taking (1 ml) from the output water and it was done with distilled water to (50 ml), then it was transferred to Erlenmeyer and the pH) of the medium was adjusted at (11), which is the degree corresponding to the conditions for the formation of the complex between the bound and the calcium ion, and the calibration began using a solution Standard of Na<sub>2</sub>EDTA at a concentration of (0.01 M), the concentration of calcium ions was calculated from the titration law expressed by the following relationship:

$$Wt_{Mg^{2+}} = 12.16 [ (total\ hardness_{(ppm)} * 0.01998) - (Wt_{Ca^{2+}}_{(ppm)} * 0.049) ]$$

where:

**Wt<sub>Mg<sup>2+</sup></sub>**: the concentration of magnesium ions in the output water in mg/L.

**total hardness:** Amount of EDTA coming down from the burette\* Equivalent weight of calcium carbonate which equates to ml of EDTA \* 1000/volume of sample used is estimated in mg/L.

**Wt<sub>Ca<sup>2+</sup></sub>**: the concentration of calcium ions in the output water in mg/L.

$$Wt_{Ca^{2+}} = 0.4008 * 1000 * V_{EDTA} \setminus V_{a\ sample}$$

**V<sub>EDTA</sub>**: Volume of standard solution consumed in ml.

**Calculation of removal yield:**

According to the removal yield (percentage of the removed quantity) from the following relationship:

$$Y\% = \frac{(C_0 - C) \times 100}{C_0}$$

where :

**C<sub>0</sub>**: element concentration before treatment with sodium phosphate (mg/L).

**C**: elemental concentration after treatment with trisodium phosphate (mg/L).

**Y%:** The percentage of the amount removed (removal yield).

**4. Results And Discussion**

**3.1 Characterization of the brine produced by reverse osmosis units:**

The following table shows the results of the analysis of samples of the incoming and outgoing water (brine solution) in the Baniyas State Company. It is noted from Table (2) that the output water resulting from the reverse osmosis units is excessively hard according to the international water classification [11].

**Table 2.** Characterization of the output water (brine)

	Input water	brine	The maximum permissible limits of the standard standards for treated water used for irrigation purposes [18]
pH	7.9-7.6	8.5-9.4	6-9
total dissolved salts (mg/L)	270	895	-
electrical conductivity (µs/cm)	550-500	2500-1698	-
Ca <sup>2+</sup> (mg/L)	150-100	520.8-320.08	400
Mg <sup>2+</sup> (mg/L)	2.4	640-533.5	60
total hardness CaCO <sub>3</sub> (mg/L)	260	2100-3000	-
SiO <sub>2</sub> (mg/L)	10	20.07-240	-

**3.2 Study of the efficiency of removing the cationic load from magnesium Mg<sup>2+</sup> using trisodium phosphate Na<sub>3</sub>PO<sub>4</sub> in accordance with the mixing time at pH=[ 8.5-9.4] and T= 20 ± 5C°:**

Figures (5, 6, 7) and tables (3, 4, 5) represent the changes in the yield of removing the cationic load from calcium Mg<sup>2+</sup> according to the mixing time dependency at the pH of the solutions corresponding to the output water pH =[ 8.5-9.4] and the temperature of the solutions T = 20 ± 5C°, when [PO<sub>4</sub><sup>3-</sup>] = 0.05, 0.1, 0.5M and solutions of magnesium ion Mg<sup>2+</sup> were prepared using magnesium chloride MgCl<sub>2</sub>:

[Mg<sup>2+</sup>]=0.005, 0.01, 0.05, 0.1M.

**Table (3):** Changes in the yield of removing the cationic load from magnesium according to the mixing time at [M 0.05]=PO<sub>4</sub><sup>3-</sup> &  $\frac{[PO_4^{3-}]}{[Mg^{2+}]} = 0.5, 1, 5, 10$

$\frac{[PO_4^{3-}]}{[Mg^{2+}]}$ time(min)	10	5	1	0.5
10	80.54%	70.1%	7%	2.5%
20	83.6%	60.2%	10%	4%



30	90.1%	68.2%	21%	10.3%
40	88.3%	68.5%	5%	12%
50	90.3%	76.6%	14%	11%
60	90%	80.3%	2%	8%

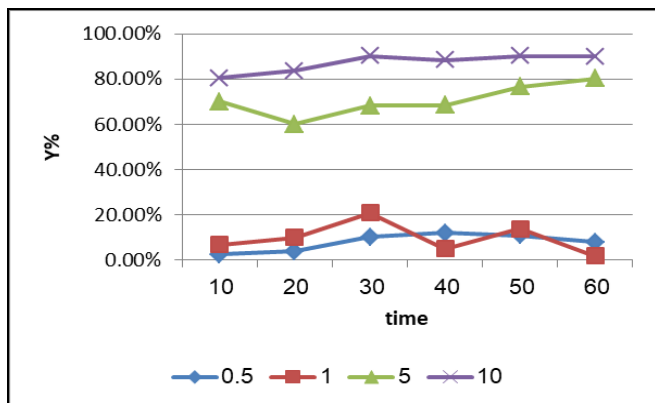


Figure (5): Changes in the yield of removing the cationic load from magnesium according to the mixing time at [M 0.05 ]= $PO_4^{3-}$  &  $\frac{[PO_4^{3-}]}{[Mg^{2+}]} = 0.5, 1, 5, 10$

Table (4): Changes in the yield of removing the cationic load from magnesium according to the mixing time at [M 0.1 ]= $PO_4^{3-}$  &  $\frac{[PO_4^{3-}]}{[Mg^{2+}]} = 1, 2, 10, 20$

$\frac{[PO_4^{3-}]}{[Mg^{2+}]}$ time(min)	20	10	2	1
10	70.9%	85%	1.1%	9%
20	75.8%	97%	4.7%	9.6%
30	70.5%	97.2%	3.3%	18%
40	80.7%	95.9%	14.2%	15%
50	95.5%	97%	10%	15%
60	92%	97.3%	7.3%	8.9%

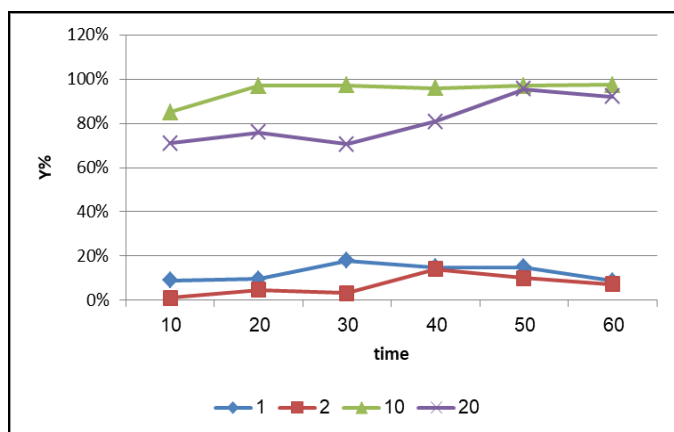


Figure (6): Changes in the yield of removing the cationic load from magnesium according to the mixing time at [M 0.1 ]= $PO_4^{3-}$  &  $\frac{[PO_4^{3-}]}{[Mg^{2+}]} = 1, 2, 10, 20$

Table (5): Changes in the yield of removing the cationic load from magnesium according to the mixing time at [ M 0.5 ]= $PO_4^{3-}$  &  $\frac{[PO_4^{3-}]}{[Mg^{2+}]} = 5, 10, 50, 100$

$\frac{[PO_4^{3-}]}{[Mg^{2+}]}$ time(min)	100	50	10	5
10	96.20%	92.0%	90.9%	55.4%
20	96.90%	92.0%	93.7%	60%
30	98.20%	95.0%	93.7%	62.3%
40	95.0%	98.0%	95.8%	50.6%
50	99.5%	98.0%	95.1%	51%
60	99.80%	97.4%	98.8%	51%

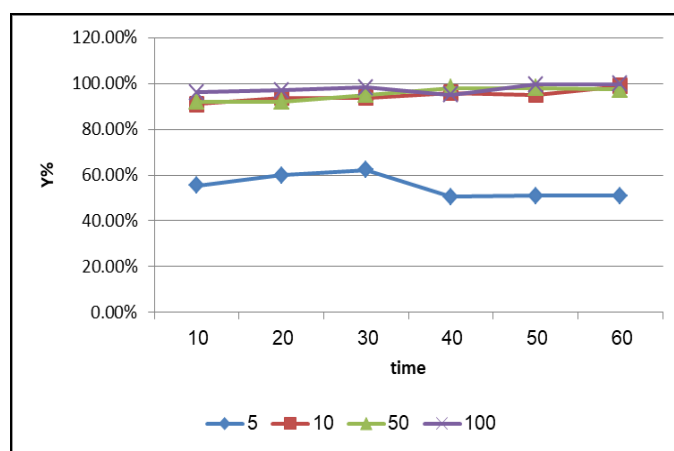
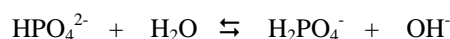
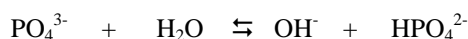


Figure (7): Changes in the yield of removing the cationic load from magnesium according to the mixing time at [ M 0.5 ]= $PO_4^{3-}$  &  $\frac{[PO_4^{3-}]}{[Mg^{2+}]} = 5, 10, 50, 100$

It is noted from figures (5, 6, 7) and tables (3, 4, 5) that to obtain the best yield it should be at the studied volumes, and that the maximum yield of precipitation  $\approx 97.3-100\%$  corresponding to the lowest possible concentration of phosphate ion  $PO_4^{3-}$  was at  $[Mg^{2+}] = 0.01M$  and  $[PO_4^{3-}] = 0.1M$ , and the equilibrium time, corresponding to maximum values of the precipitation yield of the  $Mg^{2+}$  ion, can be reached after one hour.

It is also noted that the yield of  $Mg_3(PO_4)_2$  magnesium phosphate precipitation increases with the increase in the concentration of trisodium phosphate  $Na_3PO_4$ , and this can be attributed to two main reasons. Magnesium  $Mg^{2+}$  in the form of  $Mg_3(PO_4)_2$  magnesium phosphate. As for the second reason, the increase in the sedimentation yield is associated with the phenomenon of sequential precipitation of magnesium hydroxide ( $Mg(OH)_2$ ) in alkaline media, which leads to an increase in the sedimentation yield of the magnesium ion  $Mg^{2+}$  in its phosphate and hydroxide forms according to the two equations [19]:

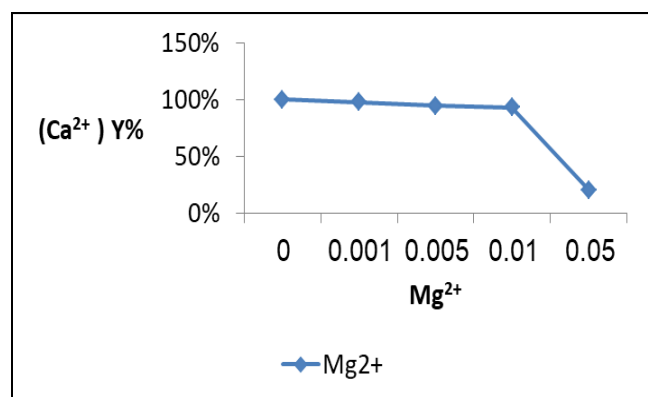


The effect of temperature decreases in increasing the precipitation yield of the  $Mg^{2+}$  magnesium ion in this case because it is one of the cases corresponding to higher concentrations of trisodium phosphate  $Na_3PO_4$ . The contradictory effect of temperature on precipitation rates can be attributed to the so-called saturation index, and an increase in temperature and concentration Phosphate ion  $[PO_4^{3-}]$  will be associated with dissolution reactions for some phosphate species, thus reducing the effect of temperature on increasing sedimentation rates [20].

### 3.3 • Study of the cationic removal efficiency of calcium according to $[Mg^{2+}]$ concentration using trisodium phosphate $Na_3PO_4$ according to mixing time at pH = [ 8.5-9.4] and $T= 20\pm 5C^o$ and mixing time of 60 minutes:

**Table (6):** Study of the efficiency of removing the cationic load from calcium according to  $[Mg^{2+}]$  concentration using trisodium phosphate  $Na_3PO_4$  according to mixing time at pH=8.5-9.4 and  $T=20\pm 5C^o$  and mixing time of 60 minutes:

$Mg^{2+}$	0	0.001	0.005	0.01	0.05
$Ca^{2+}$ (Y%)	% 100	% 97.7	% 94.3	% 93.3	% 20.7



**Figure (8):** Changes in the yield of removing the cationic load from calcium according to concentration  $M 0.1 = [PO_4^{3-}]$  and  $[Ca^{2+}] = 0.01M$  and  $[Mg^{2+}] = 0, 0.001, 0.005, 0.01, 0.05 M$

Figure (8) shows that adding magnesium ion  $Mg^{2+}$  to the studied aqueous system will lead to inhibiting the formation of calcium phosphate.

The phenomenon of precipitation of magnesium with calcium phosphate species was studied by Abbona and colleagues [21], initially forming ACP which soon turned into the more stable HAP. It is possible for HAP to incorporate a small percentage of magnesium ion  $Mg^{2+}$  into its structure, causing changes and inhibition of HAP formation [22].

In this regard, the results of some researchers suggest [23] confirm that the addition of the  $Mg^{2+}$  ion leads to the formation of a relatively unstable ACP. It has been suggested that the strong decrease of the  $Mg^{2+}$  ion in solution is due to the incorporation of  $Mg^{2+}$  into the precipitated phosphate phase  $(Ca,Mg)_3(PO_4)$ . The magnesium ion,  $Mg^{2+}$ , changed the stability of the sediment and allowed the more soluble ACP to precipitate. The addition of  $Mg^{2+}$  reduced the calcium ion  $Ca^{2+}$  precipitation rate by more than 25% [23].

### 3.4 Applying optimal conditions for removing calcium ions to the output water from reverse osmosis units:

The total hardness in the output water resulting from the reverse osmosis units was measured and amounted to (3000  $ppm$ ) and the calcium concentration was (320.08  $mg/l$ ), which corresponds to (0.0078 M), and the magnesium concentration was (533.55  $ppm$ ) taken (10 ml) from water. The output was determined by the pH and temperature, then 1 ml of trisodium phosphate was added at a concentration of (0.05 M) and at a centrifugation speed estimated at 3500 rpm, we reached a removal rate estimated at 64% for calcium ions, 60% for total hardness, and a removal rate for magnesium ions estimated at 55%. When adding (1ml) trisodium phosphate at a concentration of (0.1 M), we reached a removal rate of 100% for calcium ions, a removal rate of (95.9-97.3)% for magnesium ions, and a removal rate of ( 92.6-97)% for the total hardness, in addition to that the remaining sediment was kept for analyses. his own.

In addition, a white precipitate is formed, which is filtered and dried to obtain a white powder, as in Figure (5).



**Figure (9):** The white precipitate after filtration and drying to a white powder

## 5. Conclusion

In general, the initial treatment of the output water resulting from the reverse osmosis units resulted in the removal of (95.9-97.3) % of the magnesium ions and the removal of the total hardness by up to (95-97%). Therefore, the super-hard water that contains high concentrations of calcium ions can be converted into soft water using a phosphate substance. Sodium triglycerides.

The recommendations are limited to one recommendation, which is to work on investing the results of this research to support water resources and protect the environment, as this water can be recycled and converted into water suitable for irrigation, in addition to the phosphate residue that can be converted into fertilizer for plants.

### Data availability

Not applicable

**Conflict of interest**

The authors declare that they have no competing interests.

**Funding source**

Not applicable

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**contributions**

The authors were actively involved in the concept and design, Acquire data, interpret data and formulate manuscript as well as critical review and final approval The manuscript to be submitted.

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