Zero-Emission Water Cycle When Developing Underground Gas Storage in Rock Salt Formation

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Abstract. The aim of the work is the possibility of creating a closed water cycle during the development of underground gas storages in rock salt formations with obtaining commercial sodium chloride. Desalination experiments were carried out on standardized test solutions with various concentrations of sodium chloride (1%, 5%, and 10% solutions NaCl). Desalination was executed on a pilot electrodialysis unit, as this process has a higher degree of concentration than reverse osmosis. The dissolution rates of formation washing-out. Optimal concentrations of brine were defined experimentally. The possible salt rejection is determined. It is possible to direct the concentrate to the evaporation unit and obtain commercial salt - sodium chloride. Experiments have shown that it is possible to desalinate brine obtained by formation washing-out and reuse the desalted permeate. This reduces operational costs, as to obtain commercial product it is required smaller volume of evaporation unit. It also helps to reduce consumption of fresh water needed to salt formation washing-out. The flowsheet of the water cycle during the development of underground gas storages in salt formations with obtaining a commercial product at the output has been developed.

1. Introduction
In places of oil and gas production, pipeline transportation background, offshores that are located in difficult climatic conditions companies are implementing on a constant basis activities that require a large amount of technical water.

In areas of permafrost, thermal caverns the formation of salt water and weak brines are observed. Those are associated with cryogenic concentration (freezing) processes. In this case, water of increased mineralization may form. This is not only state of the art for the Arctic offshore and areas of the far north but also for the energy and resources market in general.

When operating transportation and storage systems for oil and gas water is required for technological equipment, for power plants, and for domestic needs. Most often, in the conditions of field development (extreme north offshore), there is water that does not meet the requirements for industrial water (salinity not exceeding 500 mg / l, pH (7-8), nitrates (2-4) mg / l, nitrites (0.03 - 0.06) mg / l, chlorides and sulfates not more than 120 mg / l). On the platform, seawater is abundant, while freshwater is scarce. Technical fresh water is delivered by tankers, which may be delayed due to weather conditions.
The use of sea water on platforms for technical needs leads to rapid wear of equipment due to corrosion, there is also a high risk of scaling, which will lead to a shutdown of the process, therefore it is necessary to use the desalination process.

To obtain fresh water that meets the requirements, it is possible to use membrane processes (in particular electrodialysis and reverse osmosis).

2. Development of underground gas storage in salt formations

Currently, the storage of produced natural gas is an urgent problem, because of the necessary to keep storage losses to a minimum. Solution of this problem is the underground gas storage in the salt formations (sodium chloride) which is produced by NaCl washing-out.

Underground gas storages in rock salt formations have a very dense structure, which leads to minimal gas losses. The salt formation has the property of self-healing cracks. Storage facilities are developed by dissolving salts.

Dissolution is carried out by supplying fresh water to the formation and dissolving sodium chloride. Fresh water can be supplied to the upper or lower part of the formation.

However, the arrangement of underground gas storages requires large amounts of fresh water and has the problem of brine utilizing. The solution to this issue is possible by desalting the brine. As a result, we obtain a diluate (fresh water with a salinity approximately 200 mg/l) and a concentrate. The diluate is directed again to the reservoir, and the concentrate is directed to the evaporator to obtain dry salt (commercial product). This allows to reduce the consumption of fresh water and get a commercial product – sodium chloride. The rate of the cavern development in the salt formation depends on the dissolution. Solubility is the limit concentration of a substance in a solution, when the solution is in equilibrium state (the chemical potential of a dissolved substance is equal to the chemical potential of a substance in a solid phase). The principal law of dissolution kinetics is formulated as follows: the specific flow of a substance per unit surface area of the dissolution is proportional to the concentration of the undersaturated solution.

The process of dissolution of many natural compounds in real hydrogeological and geochemical situations often depends on the speed of groundwater. This is due to the fact that a velocity increase is a consequence of a more active removal of dissolution products.

As a result, the concentration of a solvend in groundwater decreases, which leads to an increase in the difference in the concentration of saturation and the concentration of the real solution. Therefore, the magnitude of undersaturation increases, which is the driving force of the dissolution process. However, the mineralization of water is reduced, because an increase of the water velocity leads to a decrease in the water-rock contact time and the active removal of dissolution products. This means that the dynamics of groundwater can no longer significantly affect to the rate of dissolution of the solid phase. Therefore, the less soluble the compound, the less the groundwater dynamics affects to the kinetics of its dissolution [1-29].

Formations are mainly composed of minerals belonging to the following classes: silicates (crystalline rocks and clays), carbonates (limestones, dolomites), sulfates (gypsum), chlorides (salt rocks), which are listed in order of increasing solubility. The numerical values of their solubility differ by several orders of magnitude and usually they are divided into several groups in order to more clearly imagine the magnitude of the differences.

The lower the concentration of salts of the source water, the more efficient is the dissolution of the solid in it and the faster cavern occurs when the rock salt formation is washed-out.

Another condition that accelerates the dissolution is an increase the temperature, since this means an increase the saturation concentration determined by the solubility of the substance, as well as an increase the dissolution rate coefficient. The solubility of all compounds (except some sulfates) increases with temperature increase, sometimes significantly.

The solubility of halite (NaCl), which provides the main salt load of natural waters, varies very slightly in the range (0 – 60) °C. Therefore, the sedimentation of halite during cooling of solutions can
be expected only when rising to the surface (usually technogenic) of concentrated deep brines containing saturated solutions of chlorides.

3. The calculation of the optimal value of the brine concentration which is removed from the dissolution process

The optimal value of the final concentration at which brine should be delivered to the electrodialysis unit was calculated. In order to maintain the efficiency of the dissolution process, it is necessary not to oversaturate the solution.

For the calculation, was taken the conditional area of the dissolved salt in contact with the solution the 1 m$^2$ and the dissolution time the 1 minute. Final concentrations were considered the 1 g/l, 2 g/l, 5 g/l, 10 g/l, 50 g/l and 100 g/l.

Was accepted that fresh water with a salt concentration of 200 mg/l is supplied upon the dissolution of the rock salt formation, therefore, the difference between the saturation concentration and the initial concentration for all six cases is the same and equal

$$\Delta b_{\text{in}} = 370.8 \text{ g/l}.$$

Was calculated the difference between the concentration of saturation $C_{\text{in}}$ and the final concentration for different cases. Based on the calculated data, a characteristic curve of dissolved-solid yield versus brine final concentration was constructed (Figure 1).

![Figure 1. Characteristic curve of dissolved-solid yield versus brine final concentration.](image)

The graph shows that when removing brine with low concentrations, the mass of the salt removed is not very large. With an increase the final concentration (starting from 10 g/l), the mass of the solute also increases. The zone of effective operation of the electrodialysis unit is in the range from 2 g/l to 50 g/l. Therefore, the most optimal concentrations of brine, which should be delivered to the electrodialysis unit, are located in the interval (10 – 50) g/l. When developing an underground gas storage in rock salt formations, salt dissolves with fresh water to obtain a storage cavity. The saturated solution obtained by dissolving is called brine. When the brine is desalted, it will be divided into diluate (fresh water) and concentrate (highly mineralized residue). From the concentrate, it is possible to obtain a commercial product – sodium chloride, and the diluate can be used to re-dissolve the rock salt. As a result, it is possible to make a closed water cycle with a minimum consumption of additional fresh water and a commercial product at the outlet.

4. Experiments on standardized test solutions of NaCl

The aim of the experiment was to identify the possibility of desalination of salt solutions formed during the rock salt formation (NaCl) washing-out during the development of an underground gas storage. Since reuse of the diluate will significantly reduce the consumption of fresh water.
1% (10 g/l) and 5% (50 g/l) solutions of sodium chloride (NaCl) were prepared for the experiments. The process of obtaining fresh water was simulated for re-directing to wash-out the formation and concentrate to the evaporation plant.

The prepared solutions were passed through a pilot electrodialysis unit. Initial, intermediate and final measurements of pH, conductivity, salinity, ammeter and voltmeter readings built into the unit were carried out. Indications for the desalination of a 1% solution are summarized in table 1, and for the desalination of a 5% NaCl solution in table 2.

**Table 1.** Indications for the desalination of a 1% NaCl solution.

| Parameter            | Conductivity/ Salinity | pH | Amperage I, A | Voltage U, V |
|----------------------|------------------------|----|---------------|--------------|
| Initial solution     | 17.9 mS/cm 10 g/l      | 3  | 7.3           | 19.7         |
| Diluate after 10 minutes | 2.77 mS/cm 1.78 g/l | 6.1 | 1             | 18.2         |
| Concentrate after 10 minutes | 37.6 mS/cm 24 g/l | 7.6 | -             | -            |
| Diluate after 15 minutes | 209 μS/cm 146.3 mg/l | 5.8 | -             | -            |
| Concentrate after 15 minutes | 38.9 mS/cm 27.2 g/l | 7.6 | -             | -            |

**Table 2.** Indications for the desalination of a 5% NaCl solution.

| Parameter            | Conductivity/ Salinity | pH | Amperage I, A | Voltage U, V |
|----------------------|------------------------|----|---------------|--------------|
| Initial solution     | 73.5 mS/cm 50 g/l      | 3  | 7.6           | 19.7         |
| Diluate after 25 minutes | 24 mS/cm 16.8 g/l | 7.2 | 2.3           | 18.4         |
| Concentrate after 25 minutes | 102 mS/cm 71.4 g/l | 7.6 | 1.8           | 18.4         |
| Diluate after 42 minutes | 220 μS/cm 154 mg/l | 5.8 | -             | -            |
| Concentrate after 42 minutes | 111.3 mS/cm 77.91 g/l | 7.6 | -             | -            |

Based on the tabular data, we can conclude that it is most efficient to pass a solution with a lower concentration, because less time and energy is spent. And it is better to take less concentrated water from the washout cycle, because the lower the concentration of salts in water, the better the dissolution process.

The experiments showed that the brine obtained by washing-out the formation can be desalted and reused desalted diluate. The amount of demineralized water is 90% or more of the initial brine. It is possible to direct the concentrate to the evaporation plant and obtain commercial sodium chloride. A block scheme of water circulation during the development of a gas storage is shown in Figure 2.

The advantages of using an electrodialysis unit are: chloride ion tolerance of the membranes, a large percentage of recovery (over 90%), membrane resistance to salts and various pH.

After electrodialysis unit the concentrate is sent to the evaporation plant, where salt is evaporated. At the output, we get wet salt with a moisture content of about 70%, which goes to a centrifuge to get rid of excess moisture. After the centrifuge, salt water is returned back to the process cycle and re-fed to the evaporator with the next portion of the concentrate, and the wet salt is sent to bake. Ultimately, at the output we get dry commercial salt (NaCl).
The experiments and the block scheme showed the possibility of using an electrodialysis desalination plant when developing gas storages in rock salt formations. Using the electrodialysis process, it is possible to organize a water cycle with minimal adding of fresh water, desalinated brine (diluate) will use as the basis for the washing-out of the reservoir. The utilization of dissolution products will also be minimized, and at the output we will get a commercial dry salt of sodium chloride.

5. Utilization of brines
To maintain pressure and increase oil recovery coefficient in productive wells, the produced water is injected back into the reservoir. Along produced waters are called waters, which are formed as a result of dehydration and desalination of oil at the oil treatment unit. Part of the produced water is lost in the process of oil preparation and it is necessary to recharge water from outside to maintain balance.

There are two conditions for the formation of supersaturated solutions: an increase the actual concentration of ions and a decrease the solubility.

When water of different chemical composition is mixed, supersaturation of the resulting solution with some compound may occur. That can be the reason of the solid phase formation. Water, upon mixing which forms a solid phase, precipitating from the solution, are called "incongruent" waters. Examples of "incongruent" waters are river and sea waters, deep thermal and ground waters, deep brines and surface (river, sea waters), etc. Let us give the most typical sedimentation reactions when mixing incongruent waters:

- between calcium sulfate of sea water and sodium carbonate of groundwater, which leads to precipitation of calcite:
  \[ \text{CaSO}_4 + \text{Na}_2\text{CO}_3 = \text{Na}_2\text{SO}_4 + \text{CaCO}_3 \downarrow, \]  
- between magnesium sulfate of sea water and calcium chloride of produced water (discharge of pumped deep waters), with the formation of gypsum:
  \[ \text{MgSO}_4 + \text{CaCl}_2 = \text{MgCl}_2 + \text{CaSO}_4 \downarrow, \]  
- between sodium sulfate of river water and produced water calcium chloride (unloading of deep brines due to tectonic disturbances), with the formation of gypsum:
  \[ \text{Na}_2\text{SO}_4 + \text{CaCl}_2 = \text{NaCl} + \text{CaSO}_4 \downarrow. \]

Therefore, the main requirement for feed water is compatibility with produced water and rock. With a decrease the injectivity coefficient of injection wells from the beginning of water injection by 20%, work should be done to restore the filtration characteristics of the bottomhole zone and, if necessary, improve the quality of the injected water. Therefore, high requirements are imposed on feed water for the content of mechanical impurities.

Another important concept is carbon dioxide balance. Carbon dioxide equilibrium is the equilibrium state of a system of hydrocarbonate, carbonate ions and free carbon dioxide
CO₂ + H₂O ⇄ H⁺ + HCO₃⁻ ⇄ 2H⁺ + CO₃²⁻ (4)

It is necessary to presence of a certain amount of free CO₂ for existence of certain concentration of HCO₃⁻. It is called equilibrium (Figure 3).

Figure 3. The ratio of CO₂ and HCO₃⁻ in solution under normal conditions.

If the content of free carbon dioxide in water is more than it is necessary for equilibrium, then when such water comes into contact with CaCO₃, it will dissolve, so the reaction will go from the left to the right. The dissolution process will continue until equilibrium is reached.

The pressure, temperature, and pH changes lead to a change in carbon dioxide equilibrium, which can lead to reservoir mudding.

The deposition of carbonate salts (CaCO₃, MgCO₃) is possible in two cases:

1. Decrease the reservoir pressure and transition of CO₂ from a dissolved state to a free one. The carbonate content in the solution is mainly supported by the conditions of carbonate equilibrium, in which the partial pressure of CO₂ plays a pivotal role.

\[ \text{CO}_2 + \text{H}_2\text{O} \rightarrow \text{H}_2\text{CO}_3 \rightarrow \text{H}^+ + 3\text{HCO}_3^- \rightarrow 2\text{H}^+ + 2\text{CO}_3^{2-} \] (5)

Removing CO₂ from the system shifts the reaction equation towards the formation of CO₃²⁻ ions and the deposition of carbonate minerals. Therefore, when the reservoir pressure falls below the saturation pressure and free gas appears, conditions for the formation of carbonate sediments appear. This usually happens in production wells, when the solutions rise to the surface.

2. When water of HCO₃⁻-Na composition (inversion) is mixed with Cl⁻-Ca formation solutions, Ca²⁺ formation water interacts with the HCO₃⁻ ion of injected water, which leads to precipitation of calcite by the reaction:

\[ \text{NaHCO}_3 + \text{CaCl}_2 \rightarrow \text{CaCO}_3 \downarrow + \text{NaCl} + \text{H}_2\text{O} \] (6)

Sulfate deposition occurs when incompatible waters are mixed, usually when surface (river, sea) waters are pumped into formation brines. Due to the peculiarities of formation of low sulfates in reservoir brines (n·10 – n·100 mg/l), the content of SO₄²⁻ in seawater is 2500 mg/l and n·100 mg/l in river water. At the same time, in underground brines, with an increase in mineralization, a progressive accumulation of Ca²⁺ occurs, reaching up to (10,000 –15,000) mg/l in brines with a salinity of (200 – 250) g/l. The introduction of ”excess” sulfate ion in brines containing a lot of Ca²⁺ ion leads to the deposition of gypsum or anhydrite, depending on the reservoir temperature.

Therefore, it is necessary to adjust the feed water according to the content of sulfate and carbonate ions. For this adjustment, it is possible to use the electrodialysis process.

Using river water as an example, we conclude how efficiently the electrodialysis unit removes sulfate ions and carbonate ions. The composition of the source water, diluate and concentrate are in table 3.
Table 3. Composition of river water, diluate and concentrate after electrodialysis.

| Ion          | Source water (river) | Diluate | Concentrate |
|--------------|----------------------|---------|-------------|
| Na⁺, mg/l    | 35.2                 | 6.2     | 296.5       |
| NH₄⁺, mg/l   | 0.5                  | 0.1     | 4.2         |
| Ca²⁺, mg/l   | 230                  | 40.4    | 1936.5      |
| Mg²⁺, mg/l   | 50                   | 8.8     | 421         |
| Cl⁻, mg/l    | 230                  | 40.4    | 1936.5      |
| HCO₃⁻, mg/l  | 430                  | 75.5    | 3032.9      |
| CO₃²⁻, mg/l  | 1.7                  | 0.3     | 1.2         |
| NO₃⁻, mg/l   | 40                   | 7       | 1182.5      |
| SO₄²⁻, mg/l  | 140                  | 24.6    | 1178.7      |
| SiO₂, mg/l   | 5                    | 5       | 5           |
| CO₂, mg/l    | 1162.4               | 208.3   | 9995        |
| Salinity, mg/l | 8                  | 7.2     | 7           |
| pH           | 7                    |         |             |

The table data clearly shows, how decreased content of SO₄²⁻, HCO₃⁻, CO₃²⁻ ions. With such a composition as in the diluate, the possibility of the formation of insoluble salts when mixing feed and produced water is practically absent. Also, the reservoir will not be susceptible to mudding.

6. Conclusion

The problems of rational and efficient use of water are becoming increasingly relevant. The active expansion of the territories, especially in which mineral resources is extracted, requires new approaches to life support systems. The problem of providing fresh water in arid regions, in the Arctic, on the offshore is becoming one of the main problems that seriously affects to the economic indicators of economic activity.

Oil and gas transportation and storage systems consume large amounts of fresh industrial water. However, the development often takes place in remote areas with harsh climatic conditions, in particular: on the offshore, in arid areas, in the extreme north, where is a fresh water shortage.

This paper discusses methods for producing low-saline industrial water (salinity not exceeding 500 mg / l, pH (7-8), nitrates (2-4) mg / l, nitrites (0.03-0.06) mg / l, chlorides and sulfates not exceeding 120 mg / l) from high-mineralized water.

Electrodialysis and reverse osmosis were considered as desalination methods. The problems of obtaining fresh industrial water for offshore platforms, desalination of water before injection into the reservoir to maintain reservoir pressure and desalination of the brine during the development of underground gas storage in rock salt formations to return it to the rock salt (NaCl) dissolution cycle were considered. Laboratory-based experiments were carried out on a pilot electrodialysis unit with sea water from the Caspian Sea, with highly mineralized underground water taken from an underground source in the Beloretsky region of the Republic of Bashkortostan, model NaCl solutions of different concentrations, and on a pilot reverse osmosis unit with sea water from the Barents Sea.

Experiments have shown that electrodialysis and reverse osmosis technologies effectively desalinate water to the desired quality indicators. Also technological block schemes of the application of the electrodialysis and reverse osmosis process were developed, which make it possible to visually verify the possibility of adoption membrane processes into the transport and storage systems of oil and gas.

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