An experimental Investigation on the Impact of Brine Composition on Silica Solubility at High Temperature

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***Abstract*—** The theory of silica-water system or silica solubility is quite complex as it involves dissolution, polymerization and precipitation processes to form the silicate scale. The type and amount of silicate scale observed is dependent on several factors including pH, salinity, magnesium concentration, and the ratio of calcium to magnesium. This paper describes the impact of some factors including brine salinity and composition on silica solubility using crash quartz sandstone core samples. Synthetic brine with different salinities ranged between 20,000ppm to 60,000ppm were utilized to determine the change in soluble silica at 80°C. The amount of silica dissolved from the sandstone sample was measured using Silicomolybdate method and validated using Energy Dispersive X-RAY and X-RAY Diffraction techniques. The results clearly showed that the composition of the brine has a significant effect on the silica solubility. The amount of silica dissolution increased from 41.0mg/L to 67.8mg/L when the brine salinity increased from 20,000ppm to 60,000ppm, due to the increasing in salt contents of the brine. It was also observed that the addition of magnesium and calcium with ratio 1:1 to the brine would greatly affect the silica solubility. This has been confirmed by analyzing the samples before and after saturation by the use of Energy Dispersive X-RAY.

***Keywords*—**Silica Dissolution, Silica Solubility, Water Flooding.

I. Introduction

SILICA is a general term, which refers to silicon dioxide in all of its crystalline, amorphous, and hydrated or hydroxylated forms. It commonly implies that the silicon content is given in terms of weight of silicon dioxide (SiO_2) [1]. The common and simplest phase found in nature of silica is quartz, which is the main constituent of common sand [2], [3]. Many parts of the world including the western United States, Hawaii, Puerto Rico, Mexico, the Middle East and Southeast Asia have reported silicate scaling issues as one of the most severe problems that occurs due to silica water interactions. Silica poses several problems in all unit operations of the chemical recovery cycle i.e. evaporation, combustion and causticization [4].

Quartz is the most thermodynamically stable phase of silica [5]. The potential for silica scaling exists when the concentration of the dissolved silica exceeds the solubility limit at given temperature and pH [1], that results in the precipitation and deposition of amorphous silica, which once formed, is very difficult and costly to be removed [6]. The solubility of silica is dependent on many factors such as, pH, temperature, particle size, particle hydration and the presence of other ions like iron and aluminum [5].

In field applications; seawater –which has salinity ranged between 35,000 to 40,000ppm– is usually used for flooding operations. However, the salinity of the seawater is quite high which leads to the possibility of dissolving more silica ions. Basbar et al., [7] stated that silica dissolution ratio is increasing relatively with the increasing in brine salinity, when increased from 10,000ppm to 60,000ppm. Another fact is that the composition of the water is also affecting the silica dissolution, divalent minerals such as magnesium lead to the formation of magnesium silicate scale.

Magnesium can bridge the colloidal silicate particles and form an amorphous magnesium silicate scale [8]. The silica scaling can also be found inside the production tubes, which leads to down-hole pumps failure, operational problems as well as a defect in pipeline systems [9].

There are numerous researchers declared that silica scale formation is a highly complex process, which involves silica dissolution, polymerization and precipitation with other multivalent ions. The mechanism of silicate scale formation is very complex and poorly understood [5], [7], [10–12].

The silica dissolution starts when the injected water pH mixes with the formation water pH that presents at the reservoir. Quartz particles begin to be dissolved in the formation which results in forming the monomeric silica ($Si(OH)_3 O^- Na^+$). When the high pH water mixed with the neutral

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pH connate water near the wellbore, the dissolved silica begins to polymerize and form colloidal silica as the pH is partially neutralized.

Polymerized silica can be obtained when the concentration of monosilicic acid ($Si(OH)_4$) in aqueous solution exceeds the solubility limit of amorphous silica [13]. Then the silica scaling can be formed due to the presence of magnesium. The remaining magnesium after water softening would precipitate and interact with the neutral pH water, which causes the magnesium silicate scale.

Silica may be precipitated in one of the two ways; either by the addition of metal ions like aluminum, magnesium or calcium in soluble forms as hydroxides or oxides, or by the lowering of the pH [4]. Moreover, calcium carbonate scale can occur if the calcium is introduced to the high pH water, and it provides nuclei for the development of silicate scale [5].

II. METHODOLOGY

A series of static experiments were conducted to investigate the effect of brine salinity and composition on silica solubility from a crashed sandstone sample. For each test, six grams of crash core were saturated into 30 mL. The samples were kept for 3-5 days in the oven at 80ºC until total equilibrium was achieved. The soluble silica was measured using the Silicomolybdate method with chemical reagent packets at 80ºC [14]. The detection of the soluble silica concentration was obtained using Spectrophotometer DR/2800. To validate the above measurement, Energy Dispersive X-Ray (EDX) was used to estimate the percentage of silica present in the sandstone samples before and after saturation with the brine solution.

III. RESULTS AND DISCUSSION

A. *Basic Core Identification*

The main components present in the core sample were identified by using XRD and EDX techniques. Analytical instruments such as Scanning Electron Microscope (SEM) and X-ray diffract-meter (XRD) are widely used in the area of materials research [15].

The basic core mineralogy obtained by XRD is demonstrated in Fig. 1, which shows that quartz is the main dominant component of the core sample. As illustrated in Fig.1, the grains size is not varied due to the constant temperature, as the size of quartz grains decreases with extreme increase in temperature and pressure [2], [16]. Three crystal planes were presented in the sample, quartz low-alpha, quartz alpha and quartz SiO_2. All samples have the same d-spacing value of 1.5406 A° which is the distance between atomic in the crystal. All samples have a hexagonal structure and the lattice parameters were also similar. Table 1 summarize the crystallite structure for the used sand grains.

The results obtained from EDX for the sample confirmed the presence of the quartz as the highest component (Fig. 2). The X-ray spectrum of the sample indicated that additional minerals such as aluminum oxide (Al_2O_3) and Pyrite (FeS_2) were present but with lower concentrations as compared with the quartz. The concentration of each component obtained from EDX is tabulated in Table 2, with total silica concentration of 96.31%.

S1

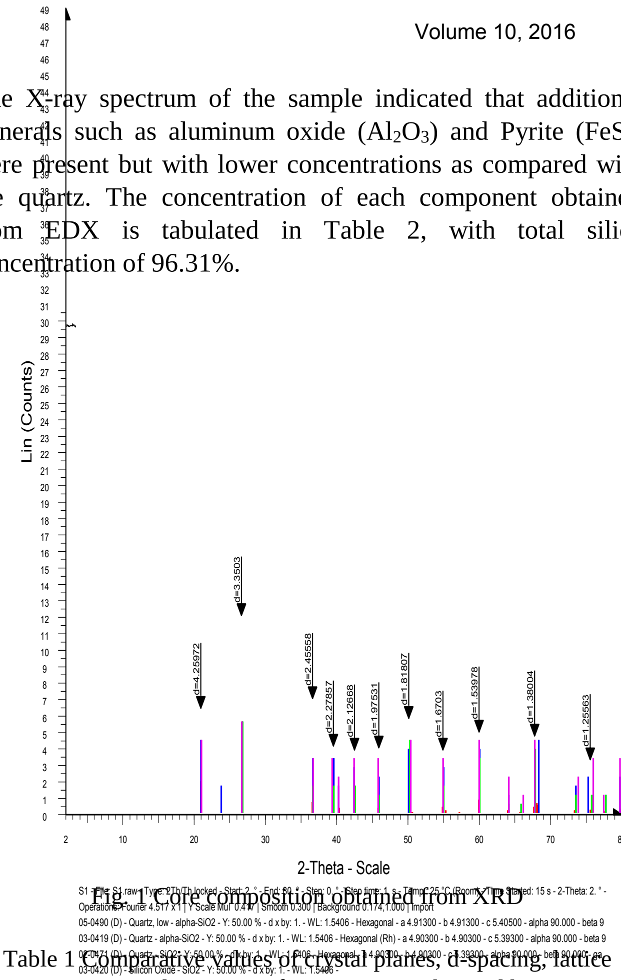

Fig. 1 Core composition obtained from XRD

Table 1 Comparative values of crystal planes, d-spacing, lattice parameters and structure of quartz grains obtained by XRD

X-Ray Diffraction			
Sample	d-spacing A°	Lattice parameter	Structure
Quartz, low-alpha SiO_2	1.5406	a = 4.91300 b =4.91300 c = 5.40500	Hexagonal
Quartz, alpha SiO_2	1.5406	a = 4.90300 b =4.90300 c = 5.39300	Hexagonal
Quartz, SiO_2	1.5406	a = 4.90300 b =4.90300 c = 5.39300	Hexagonal

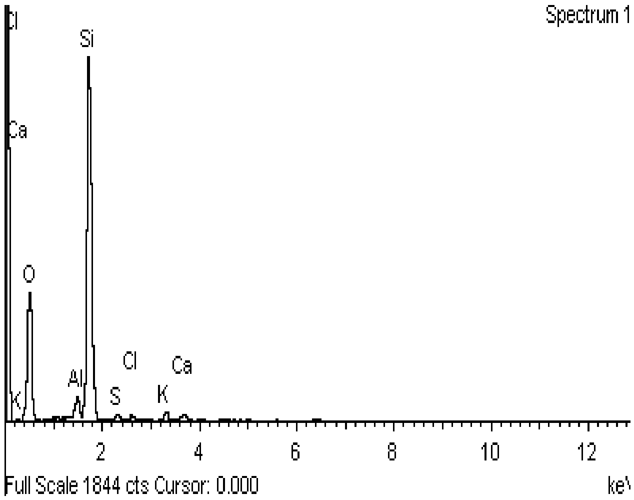

Fig. 2 Main core components obtained from EDX

Table 1 Basic core component concentration obtained from EDX

Component	Concentration %	Component	Concentration %
Silica (SiO_2)	96.31	Calcium Oxide (CaO)	0.43
Aluminum Oxide (Al_2O_3)	1.50	Potassium oxide (K_2O)	0.65
Pyrite (FeS_2)	0.61	Ferric oxide (Fe_2O_3)	0.49

B. Brine Salinity Test

The effect of brine salinity on silica dissolution was investigated using distilled water and three samples of synthetic soft brine with salinities ranged between 20,000 to 60,000ppm at 80°C. All samples were prepared using sodium chloride. Fig. 3 shows that the silica dissolution increases continuously as the brine salinity was increased. The silica dissolution was increased from 41.0mg/L to 67.8mg/L when the brine salinity was increased from 20,000ppm to 60,000ppm respectively. This is mainly due to the effect of degree of hydration [5]. When the crystalline silica grains contacted with the water, they contain this water inside the crystals, which known as the water of hydration. The amount of this water decreases with the increasing of salt contents, meaning that the high hydrated particles have low silica solubility which have the low salt contents. These results are in agreement with Gill [5].

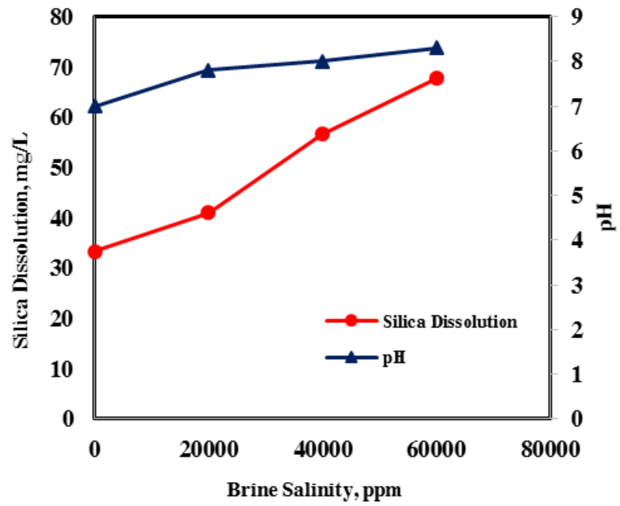

Fig. 3 Brine salinity effect on silica solubility at 80°C

C. EDX Analysis

EDX technique was employed to further confirm and provide supporting evidence of the silica solubility measurements obtained from the crash sandstone samples. For that purpose, two slides of core samples named A-F.1 and A-F.4 were placed under EDX analyzer before and after saturation at 80°C. Sample A-F.1 was saturated with distilled water while sample A-F.4 was saturated with 60,000ppm brine solution. Silica dissolution ratio (SDR %) was calculated based on the silica contents in the samples before and after saturation. Based on the obtained results, silica contents was reduced by 0.51% in sample A-F.1 when distilled water was used. However, a significant reduction of silica contents was observed in sample A-F.4 indicated a reduction by 7.32% due to the presence of salt contents in the 60,000ppm brine. This reduction in silica concentrations in both samples is referred to the silica water interactions. Table 3 shows the obtained from EDX Analysis spectrum.

D. Brine Composition Test

The effect of brine composition on silica dissolution was investigated using various concentrations of Magnesium (Mg^{2+}) and Calcium (Ca^{2+}) with ratio of 1:1 to be added to the 40,000 synthetic brine solution at 80°C as illustrated in Fig. 4. Results obtained from this test indicated that the presence of Magnesium and Calcium ions have a significant impact on increasing the soluble silica amount.

The silica dissolution was increased from 56.6 mg/L to 98.3 mg/L due to the presence of 50ppm of Magnesium and Calcium each. This is due to the massive depletion of the silicic acid by hard minerals, which results in dissolving more silica as agreed with Basbar et al. [7]. However, further increasing in silica dissolution was observed when Magnesium and Calcium concentrations increased continuously.

Table 2 Silica Dissolution Ratio (SDR %) obtained from EDX analysis, at 80°C

Sample ID	Brine Salinity, ppm	Silica Concentration Before Saturation, %	Silica Concentration After Saturation, %	SDR, %
A-F 1	0	96.29	95.80	0.51
A-F 4	60,000	96.31	89.26	7.32

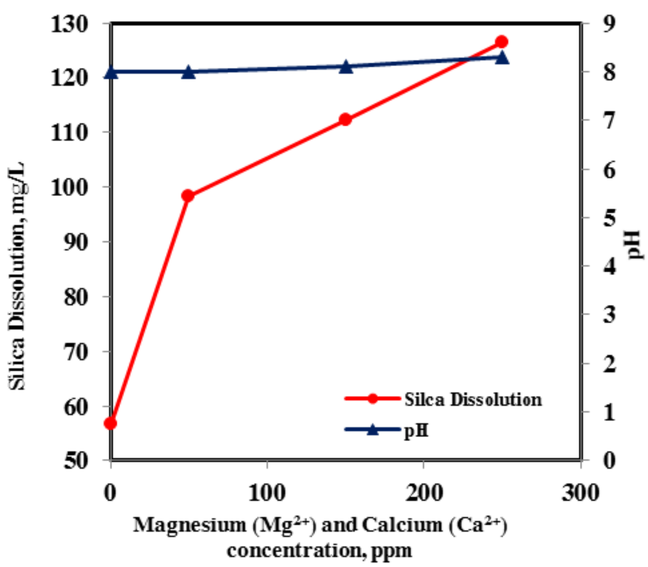

Fig. 4 The effect of Mg2+ and Ca2+ ions on silica dissolution using crash core samples at 80°C

IV. CONCLUSION

The presented results in this paper indicate that the brine salinity and composition have a significant effect on silica solubility. Increasing the amount of salt contents in the brine is the main cause of silica dissolution due to the effect of degree of hydration. Silica dissolution measurements were validated using EDX technique. A reduction of silica contents by 0.51% and 7.32% was observed when distilled water and 60,000ppm soft brine were used respectively. The presence of Magnesium and Calcium ions encountered a significant increasing in silica dissolution, which refers to the considerable depletion of the silicic acid by hard minerals.

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