



The Influence of Kaolinite and pH on Permeability in the Zubair Reservoir in the North Rumaila Oilfield, Southern Iraq

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Abstract

This research involves the study of permeability declination as a result of kaolinite due to the changing in pH in the Zubair reservoir (Lower Cretaceous) during the secondary production by water injection method. Four wells and six core samples within the North Rumaila field are studied, Minerals have been diagnosed by XRD and this specific site of clay minerals was diagnosed within the core samples by scanning electron microscopy (SEM). The core samples are also studied petrographically using the polarizing microscope and found that they mainly consist of quartz, while the predominant clay is kaolinite. The effect of pH on the values of permeability was examined through a series of laboratory experiments, as it has been tested in the cases of gradual sudden increase form acidic to alkaline. Petrophysical properties (porosity and initial permeability) were measured a prior to testing. After performing these tests, the final permeability was also measured as well as the rate of formation damage. The final permeability decreased gradually at a rate of 20-30% M Darcy during the injection out with solution of pH 3 to 11 with getting formation damage up to 25%. While in the case injection with sudden increase pH from acid to alkaline directly, there has been a rapid and substantial reduction in the final permeability as average of 28% -72%, with a formation damaged rate of 44%. The results confirmed that the reason of the decrease in the permeability is due to the kaolinite mineral which is subject to the dispersion phenomenon during the change in pH, and the acidic environment is suitable for the reservoir, and does not lead to a reduction in permeability.

Keywords: Zubair reservoir, pH, Kaolinite, Permeability, Formation damage.

تأثير الكاولينايت والذالة الحامضية على النفاذية في مكن الزبير النفطي ضمن حقل الرميلة الشمالي،

جنوب العراق

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المستخلص:

يتضمن هذا البحث دراسة تأثير معدن الكاولينايت والذالة الحامضية على قيم النفاذية في مكن الزبير (الكريتاسي الاسفل) أثناء الانتاج الثانوي بواسطة اسلوب الحقن بالمياه. تم دراسة أربعة آبار ضمن حقل الرميلة الشمالي، وست عينات صخرية (لباب صخري). شخصت المعادن بواسطة تقنية الاشعة السينية الحائدة

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(XRD) وكذلك تم تشخيص موقع الكاولينايت في العينة اللبائية بواسطة المجهر الإلكتروني الماسح (SEM)، تم دراسة العينات بتروغرافيا بواسطة المجهر المستقطب. وجد ان العينات اللبائية تتكون اساسا من معدن الكوارتز، بينما كان المعدن الطيني السائد هو الكاولينايت. تم اختبار تأثير الدالة الحامضية على قيم النفاذية من خلال مجموعة من التجارب المختبرية. اذ تم اختبار تأثير الدالة الحامضية في حالتي الزيادة التدريجية والمفاجئة من الحامضية الى القاعدية. تم قياس الخصائص البتروفيزيائية كالمسامية والنفاذية الأولية قبل اجراء التجارب. بعد تنفيذ التجارب، تم قياس النفاذية النهائية والتضرر الطبقي. النفاذية النهائية تناقصت تدريجيا بمعدل يتراوح من 20-30% ملي دارسي خلال الحقن التدريجي بالمحاليل القاعدية من 3 الى 11 مع حصول تضرر طبقي يصل مقداره الى 25%. بينما في حالة الحقن بالمحاليل مع زيادة مفاجئة للدالة الحامضية، فقد حدث انخفاض سريع وكبير في النفاذية النهائية تراوح معدله من 28%-72%، مع حدوث تضرر طبقي بلغ مقداره 44%. اكدت النتائج ان السبب في انخفاض النفاذية يعود لمعدن الكاولينايت حيث يخضع لظاهرة الانتشار والهجرة خلال تغير الدالة الحامضية مما يسبب في تراكمه حول اعناق الفتحات المجهرية مسببا غلقها. وان البيئة الحامضية هي بيئة مناسبة للمكمن ولا تؤدي الى حدوث انخفاض في النفاذية.

Introduction

Permeability is considered one of the most important reservoir specifications controls the oil production. Reductions of permeability have been observed in many global oil reservoirs [1]. The presence of clay minerals is one of the reasons affecting the permeability.

These minerals are exposed to swelling or dispersion, in case of change of the chemical environment of the reservoir as a result of water injection with chemical specifications differ from the specifications of the oilfield water in reservoir, and thus influence on reservoir performance and production rate.

Water injection is a common method in oil fields for the purpose of pressure maintenance and enhanced oil recovery. A series of experimental works has been conducted to study the decrease in permeability of the Zubair reservoir resulting from clay minerals due to change in hydrogen number (pH).

Location and description of the study area

The North Rumaila Field is located about 54 km west of Basra City. This oil field is a double plunging, simple and slightly asymmetrical anticline, with the long axis trending N-S. It is 40 km long and 13.5 km wide. Four wells are selected for collecting the core samples figure-1.

All Cores are sandstones. The Zubair Formation is divided into five members in North Rumaila field. These were informally named from top to bottom Upper Shale Member, Upper Sandstone Member, Middle Shale Member, Lower Sandstone Member, Lower Shale [2]. The subdivisions are based on the dominance of sand on shale in the lithofacies of the members.

The current study concerns the Upper Sandstone Member which is generally composed of clean friable porous sandstone, intercalated with few thin shales and fewer siltstone layers. It has been deposited in a moderate fluvial-dominated deltaic environment. Diagenetic processes (mainly cementation) are partially lithified the clastic sediments giving the rock its friable nature [3].

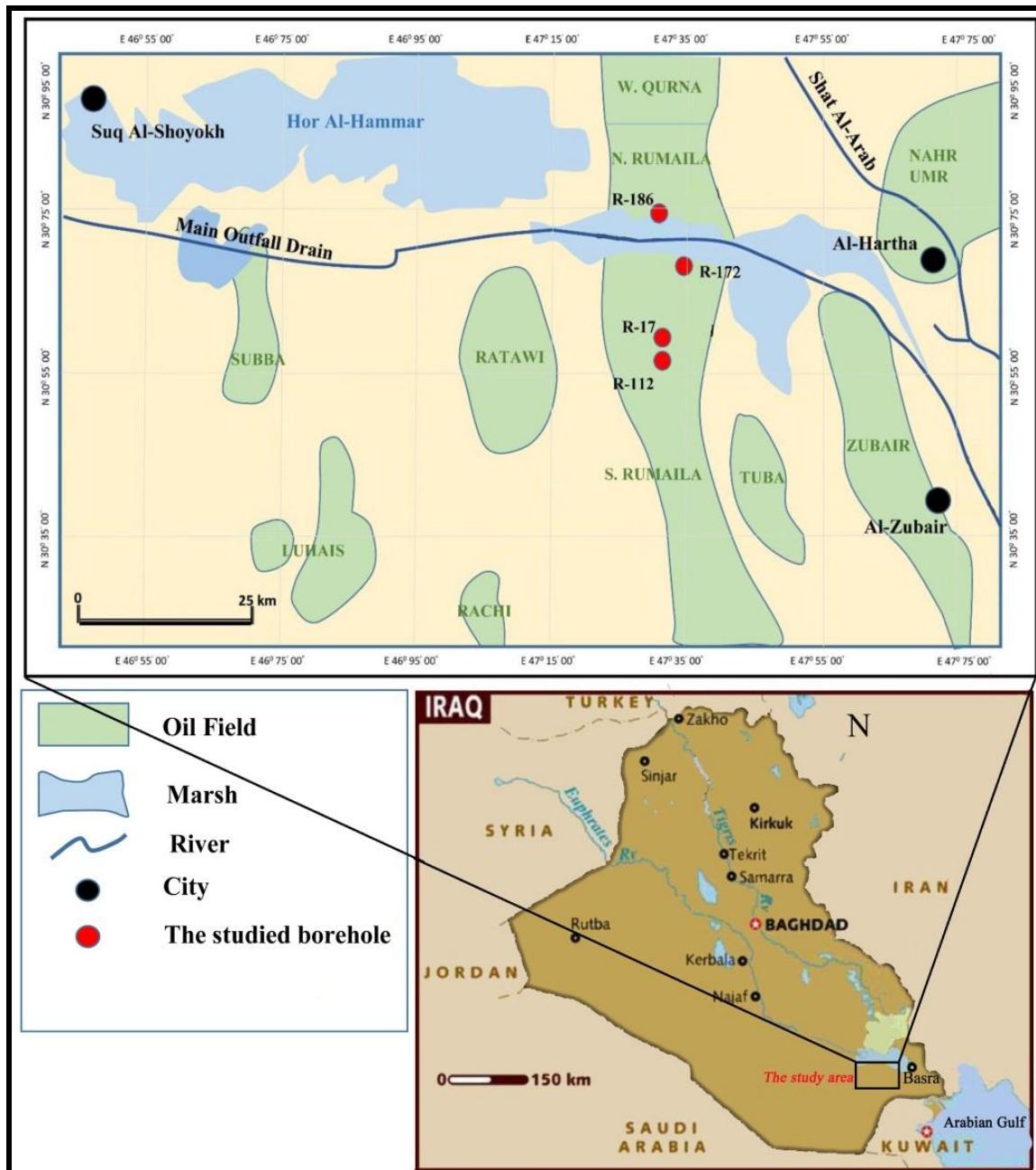


Figure 1- Location map shows the study area (North Rumaila field).

Materials and methods

Six core samples are collected from four wells in the North Rumaila Field. These wells are R-172, R-17, R-112 and R-186. The minimum and maximum depths are 3050 and 3185m respectively table-1. Core samples are subjected to XRD at the Iraq Geological Survey to identify the mineralogical composition and the type of clay minerals as well as to the SEM at the Central Laboratories of South Oil Company to determine the site of kaolinite accumulations within the cores. Rocks components are examined by XRD bulk sample method whereas clay minerals are studied (normal, heating and ethyl glycolation) according to standard procedure of [4,5]. Petrographic study using polarized microscope was also conducted on many thin sections that are prepared in the workshop of the Department of Geology, College of Science, University of Baghdad. Petrophysical analysis and core flooding are prepared at the Central Laboratories of South Oil Company. The core samples (plugs) with 3 inch length and of diameter 1.50 are cleaned from Hydrocarbon by sox let extraction using a toluene as organic solvent. To ensure a good cleaning, a Hexane material as another organic solvent is used then the core samples are dried in an oven at 60° C for 6h. Petrophysical analyses included measuring the

porosity, air permeability and liquid permeability are measured. Many laboratory experiments of core flood were conducted where the cores have been injected with brine solution of NaCl. However, Formation damage ratio of permeability is calculated according to the following equation:

$$Dr = 100 - \frac{K_{Lf} \times 100}{K_{Li}}$$

Where: Dr = Damage ratio,

K_{Li} = Initial permeability

K_{Lf} = Final permeability

A saline solutions of sodium chloride are prepared in the laboratory with different pH (strong acid 3- strong alkaline 11) to inject into the plug samples for the purpose of calculating the damage in permeability.

Table 1- Number and depth of the core samples (RC) of Zubair reservoir in the North Rumaila Field.

| Well No. | Depths (m) | Core Sample (RC) |
|----------|------------|------------------|
| R-186 | 3185 | 1 |
| R-172 | 3050 | 1 |
| | 3045 | 1 |
| R-17 | 3120 | 1 |
| | 3125 | 1 |
| R-112 | 3155 | 1 |
| Total | | 6 |

Results and discussion

The influence of kaolinite on permeability

XRD and SEM test shows that the clay minerals are represented by kaolinite figure-2. Kaolinite is formed mainly by decomposition of K-feldspars, granite, and aluminio-silicates. It is widespread in sedimentary rocks. It has ability to migrate its site filling the blanks, which it a negative impact on porosity and permeability of the oil reservoir [6]. The crystalline structure consists of 1:1, one layer of the mineral consists of an alumina octahedral sheet and a silica tetrahedral sheet that share a common plane of oxygen atoms and repeating layers of the mineral are hydrogen bonded together [7]. As a consequence of this structure, the silica/oxygen and alumina/hydroxyl sheets are exposed and interact with different components in the solution [5]. XED test for the bulk samples revealed that the samples are mainly composed of quartz, while the clay fraction is formed from kaolinite figure-2.

Petrographic study has been made in thin sections prepared from the hard core sandstones plate, 1 collected from different depth of oilfield drilled in the North Rumaila Field. In sandstone cores of the Zubair reservoir, Quartz is the most abundant grain type. Two patterns of bitumen in the hand specimen cores and under polarized microscope are observed; bitumen filling fractures, and bitumen impregnation indicating a high porosity. The important diagenetic changes that have been observed are grain compaction leading to pressure solution effect, authigenic development of minerals and sometime overgrowths, cement of silica and clay in most does not cover the whole sample. Dissolution in reservoir of acidic nature is almost dominant and contributes to enlarge the gaps, voids, cracks, fissures and fractures. In Some places of cores, very fine particles are forced and migrated by oilfield water or crude oil itself from place to another causing closure of the opening pores and eventually reducing permeability.



Plate 1- Core Sample (RC-17, Depth = 3157m), (RC-184, Depth = 3199m) collected from the Zubair Formation in North Rumaila Field

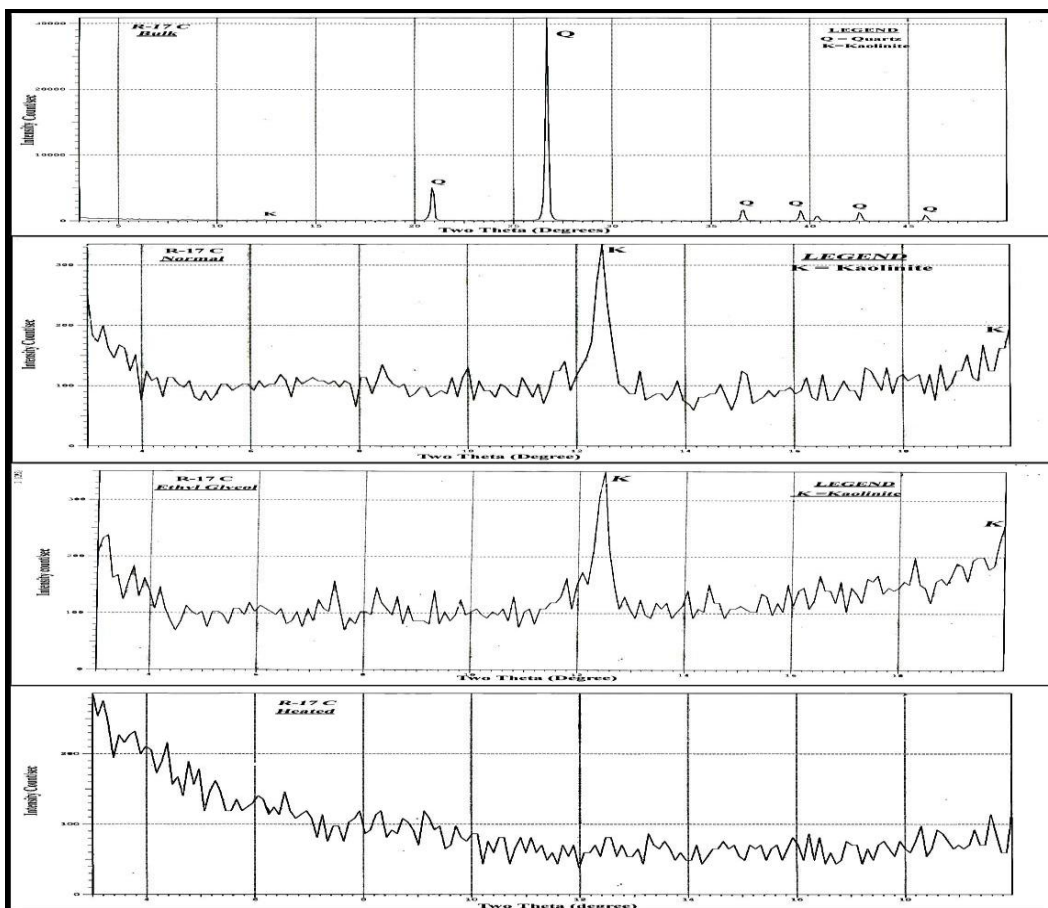


Figure 2- X-Ray diffractogram of sandstone core sample (well no R-17C; depth 3157m).

Kaolinite is one of the clay minerals, which have a balancer charge or container charge very few, this charge is centered on the outer surface of the mineral and is affected by pH, and created as a result of hydration the bonds which linking Si-O, Al-OH formed hydroxyl ion (OH) on the mineral surface [8]. Typically, kaolinite has a cation exchange capacity (CEC) of 3 to 15 meq/100 g [5], while values quoted for the specific surface area of kaolinite are from 10 to 20 m²/g [9]. The cation exchange capacity (CEC) of kaolinite strongly depends on the particle size (both thickness and diameter in the 001 plane), and pH value. Particle size is more important than crystallinity in affecting kaolinite CEC. It is commonly believed that cation exchange occurs due to the broken bonds around the crystal edges,

the substitutions within the lattice, and the hydrogen of exposed surface hydroxyls that may be exchanged [10]. Based on the above, [8] the pH value of solution smaller than zero point charge (ZPC) of kaolinite, mineral surface will be rich in hydrogen ion (H^+) with a positive charge. Thus the surface of the mineral will be charge positive as well; but if the value of pH greater than the value of ZPC of kaolinite, the mineral surface is rich in hydroxyl ion (OH^-) with a negative charge. When the pH value is equal to the value of ZPC, the surface of the mineral charge will be positive, because the amount of ZPC to kaolinite is within the limits of the acid. Generally, value of the ZPC of the kaolinite ranging from 2 - 4.6. The SEM study displays that the clay mineral grains have undergone partial pressure resulting in the formation of deterioration the reservoir quality and shows the commonly occurring clay mineral in the sandstone of Zubair reservoir is kaolinite which is wide occurrences in stacking of book pattern around inter-granular pore, this leads to narrowing these pores. Authigenic growth of kaolinites reduces the permeability/porosity ratio figure-3a .

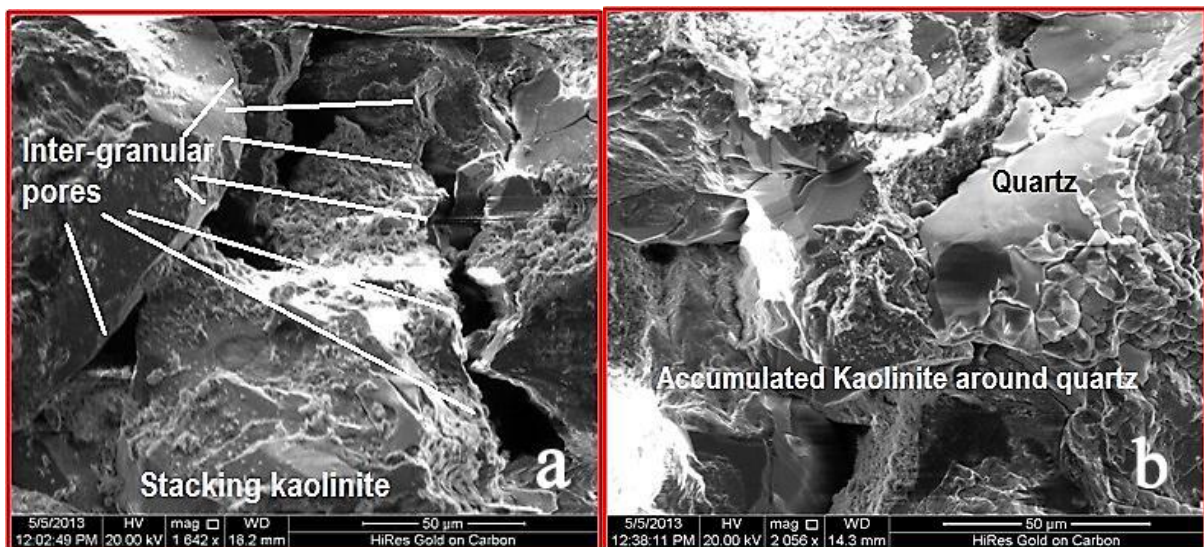


Figure 3- (a) SEM image displays how kaolinite stacking around inter-granular pores. Sample no R-17C, (b) SEM image display quartz grain buried by accumulated kaolinite (sample noR-17C).

Also kaolinite occurs around quartz grain because authentically generated from feldspar transformation and from very fine grains of kaolinite migrated from its original place to other place and accumulated in large quantities reducing permeability figure-3b. This case named fines invasion and migration. Clay migration can cause severe permeability reduction even in so-called "clean" sandstones. This type of damage can drop permeable sandstone to less than 1% of its original permeability in a few hours or less of flow.

Influence of pH on permeability

For the purpose of knowing the permeability decrease in the Zubair reservoir, brine solutions with chemical specifications similar to the formation water in terms of the (salinity and pH) has been prepared. The range of core porosity is 14 to 20% and of initial permeability is varied between 90 to 160 M.D. The cores (plugs) are saturated and injected with solutions of variable hydrogen number (pH) from strong acidic (pH=3) to strong alkaline (pH=11). These tests were carried out by two main stages: gradual and sudden stages:

Gradual increase in pH

The gradual increase in pH is defined as increase the pH from 3, 5, 7, 9 and then 11 step by step. The initial permeability and final permeability for solutions of different stages is listed in table-2. Formation damage for each test was computed based on the initial and final permeability table-2. The results shown in table-2 revealed a clear reduction in the final permeability (KL_f) during the gradual increase in the pH value from acidic (3 to 5), neutral (7) and then to alkaline (9 to 11). As example the sample RC-184, initial permeability (KL_i) was 105 M.D, after a gradual increase of pH, it declined to 80 when pH was 11, and the value of the formation damage became 24%. In all these experiments, the permeability was decline when solution goes towards alkaline figure-4. This means that the cores of the Zubair reservoir produce dispersions of native clay (kaolinite) during core flood tests, so the

permeability declining was observed. Such this conclusion has also been found [11]. These results suggest that the increasing alkalinity of the solution leads to increase the formation damage figure-5.

Table 2- Initial, final permeability, porosity and formation damage during the gradual increase of pH injected to the core samples of Zubair reservoir (North Rumaila Field).

| $\Delta\%$ Permeability | pH=11 | pH=9 | pH=7 | pH=5 | pH=3 | KL_i (M.D*) | \emptyset^{**} (%) | Depth (m) | Core No |
|----------------------------|--------------|------|------|------|------|-------------------------|-------------------------|--------------|---------|
| | KL_f (M.D) | | | | | | | | |
| 25 | 80 | 90 | 97 | 103 | 104 | 105 | 14 | 3185 | R-184C |
| | | | | | | Formation damage (Dr %) | | | |
| 25 | 75 | 77 | 90 | 100 | 102 | 100 | 14 | 3120 | R-17C |
| | | | | | | Formation damage (Dr %) | | | |
| 20 | 70 | 70 | 82 | 90 | 100 | 90 | 10 | 3155 | R-112C |
| | | | | | | Formation damage (Dr %) | | | |
| 30 | 130 | 135 | 140 | 160 | 160 | 160 | 20 | 3050 | R-172C |
| | | | | | | Formation damage (Dr %) | | | |

\emptyset = porosity; KL_i = Initial permeability; KL_f = Final permeability.

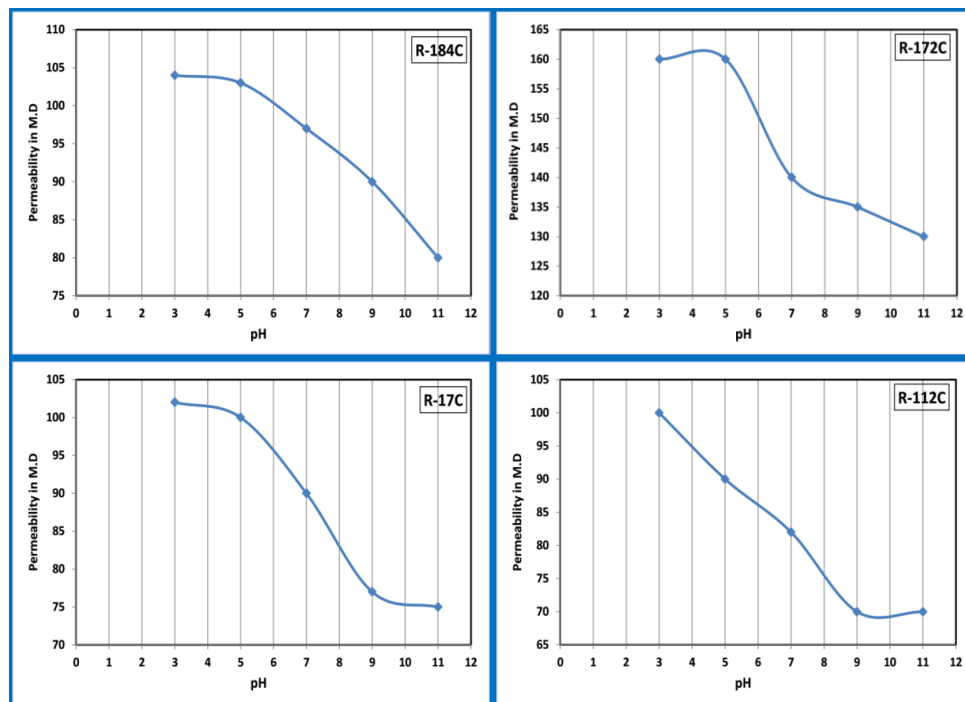
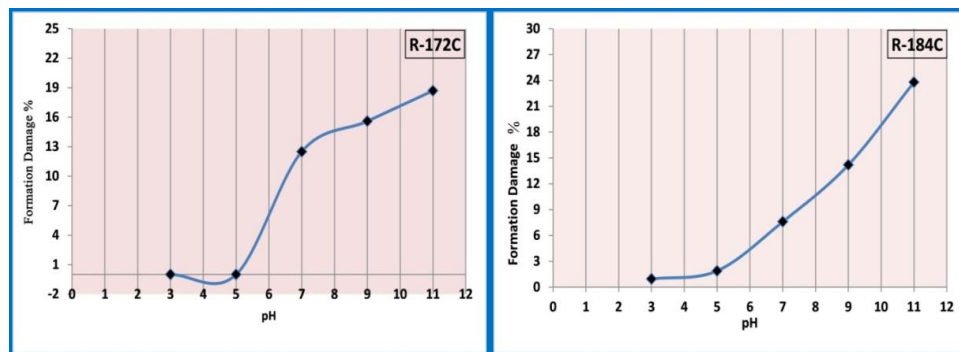


Figure 4- The negative relationship between permeability and pH during the gradual change in pH (core flood) for 4 core samples collected from 4 wells of the Zubair reservoir (North Rumaila Field).



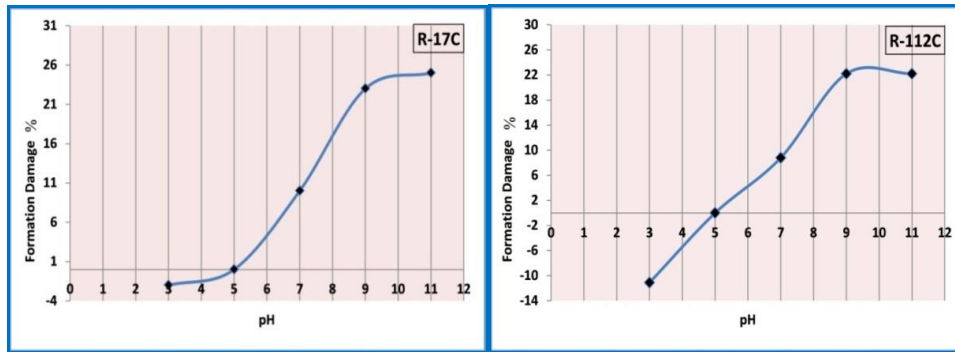


Figure 5 - Formation damage during the gradual change in pH (core flood) in 4 samples collected from 4 of the Zubair reservoir (North Rumaila Field).

Kaolinite subjects to the dispersion phenomenon and leads to partially filling of the pores [12,13]. Accordingly, the declining in permeability is caused by kaolinite during change the pH. Reason can be explained in terms of who gets absorption of sodium ion (Na⁺) on the surface of kaolinite, where it gets ionic exchange between the sodium ion and hydrogen ion (H⁺) in the solution, this leads to an increase concentration (OH) in solution and thus increases the value of pH [14]. [11] Confirmed emphasized the possibility of damage from migrating clays and the degree of damage is believed to be a function of the amount of clay minerals . The presence of small crystals of the kaolinite figure-2 may be the reason of damage.

Sudden increase in pH

Core flood test was carried out on 2 core samples. These samples were injected with brine solution of NaCl with sudden changing in pH. The results of these experiments are listed in table-3. This test revealed a sharp decline in permeability where it declined in the core sample R-17C from 72 M.D to 39 M.D with formation damage of 44.3%, whilst in core sample R-172, it declined from 165 M.D to 100 M.D with formation damage of 39.3%.

Table 3 - Resultsof permeability and formation damage under the sudden rising of pH from acidic to alkaline of samples collected from 2 wells of the Zubair reservoir (North Rumaila Field).

| Δ% Permeability | pH=11 | pH=3 | KL _f (M.D*) | Ø** (%) | Depth (m) | Core No. |
|-----------------|-----------------------|------|-------------------------|---------|-----------|----------|
| | KL _f (M.D) | | | | | |
| 32 | 39 | 72 | 70 | 8 | 3125 | R-17C |
| | 44.28 | -2 | Formation damage (Dr %) | | | |
| 68 | 100 | 165 | 165 | 18 | 3045 | R-172C |
| | 39.3 | 0 | Formation damage (Dr %) | | | |

The sudden increase has more influential than the gradual increase. Generally, the results show that these cores get damaged substantially by change in pH. The increase in damage is due to exchange of hydrogen ions in water with adsorbed sodium ions. [15] Explained that Van Der Waals attractive forces and electrostatic repulsive forces between the surface and the clay particles. The concentration of salt solution and pH affect on the repulsive forces. The surface potential of kaolinite is influenced by type of cation and the pH, causing the critical salt concentration values of sodium to be different. The damage of Zubair Formation happens by clay dispersion and decline permeability. This is considered to be the best available evidence of clay migration damage as opposed to clay swelling damage, because of the Kaolinite is the only clay mineral dominant. Generally, the effect of gradual increase of pH produced a decline in permeability reach to (20-30%) table-2, whereas during sudden increase in pH the permeability declined with average from (32- 68%) table-3. Therefore, the sudden increase is more damage from gradual increase of the Zubair reservoir rock.

Conclusions

Significantly, the principal patterns in clay migration damage have been identified by X-ray diffraction and SEM as tiny hexagonal kaolinite crystals invaded the intergranular pores and stacked

around quartz grain. These hexagons are dislodged from the pore walls in sandstones and are trapped in pore throats where they build up an internal filter cake within the sandstone. The dispersion is created as a result of clay-water reaction from dissolved species, which that yields clay hydration and swelling, or clay particle dispersion and pore plugging by micro migration with produced or injected water. [16] mentioned like this discussion. The core samples of Zubair reservoir produces dispersions of kaolinite during core flood tests, while permeability declines. The mechanism most reasonable to clay dispersion is that a certain proportion of the small kaolinite crystals grow on the surfaces of other minerals such as quartz [11]. The reason is the weak Van Der Waal forces and addition to the attract force in solution. Since the Zubair Formation is of Late Cretaceous and consists mainly of sandstone, siltstone, and shales, the authigenic clay present kaolinite is the most abundant. So, the phenomenon of clay dispersion is clear evidence of the damage in this reservoir. The loss of permeability of sandstones can be attributed either to clay swelling in the rock pores and clay particle migration. Since the prevailing clay mineral is kaolinite, so prefers likely migration to swelling. The degree of damage is believed to be a function of the amount of kaolinite that has been migrated. In Zubair reservoir there is no swelling mineral like montmorillonite, this was revealed by the XRD and SEM. Kaolinite breaks apart into fine particles. the only the finest size crystals of authigenic kaolinite are mobile. kaolinite crystals grow on the surfaces of other minerals such as quartz in the sandstone of Zubair Fromation. These clay crystals would be held in place only by weak Van Der Waal forces and the attraction of bonds shared by doubly charged ions such as calcium under the the pH influence. Another potentially significantly damaging mechanism of permeability impairment in sandstone reservoirs containing oils is that of clay deflocculation.

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