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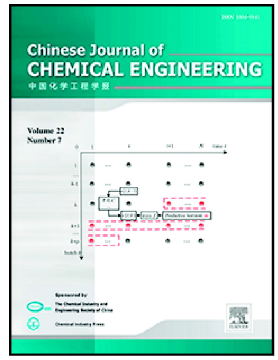

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Experimental Investigation of Different Brines Imbibition Influences on Co-and Counter Current Oil Flows in Carbonate Reservoirs

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Abstract

Imbibition of water, as wetting phase in oil-wet fractured carbonate reservoirs, plays a key role in fluid flow between matrix and fracture system. The type of injected seawater and its chemistry would profoundly influence the imbibition process. In this study, the impact of smart water (a brine that its ions have been adjusted to facilitate oil recovery) and low salinity water on co- and counter-current imbibition processes for oil-wet carbonate cores has been experimentally investigated.

The results show an increase of about 10% in oil recovery for co- and counter currents for smart seawater imbibition compared to that of low salinity seawater. In addition, as a result of the influence of co- and counter current on each other, by co-current removal from one core face, the counter current in the other face would be intensified by as much as about 75%. A close examination of different lengths (5, 7 and 9 cm) of carbonate cores with the same permeability revealed that by decreasing porous medium length, the amount of counter current producing oil would be decreased so that in the 5 cm core, counter current oil production will not happen. For similar core lengths by increasing permeability, the share of counter current flow would be decreased approximately 18% since the capillary pressure could not overcome non-wetting phase viscous forces. Considering the role of matrix length along with a modified brine (which is designed according to the matrix mixture) strengthen the relevant mechanisms to have more oil production so that the higher thickness of matrix causes the higher amount of co-current oil producing and consequently more total recovery.

Graphical abstract

Keywords: Smart water, Co-current, Counter current, Wettability alteration, Oil recovery

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Keywords: Smart water, Co-current, Counter current, Wettability alteration, Oil recovery

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Acronym	
EOR	**E**nhanced **O**il **R**ecovery
CA	**C**ontact **A**ngle
IFT	**I**nter **F**acial **T**ension
WA	**W**ettability **A**lteration
WAI	**W**ettability **A**lteration **I**ndex
CBP	**C**apillary **B**ack **P**ressure
XRD	**X**-**R**ay **D**iffractometry
SARA	**S**aturates, **A**sphaltenes, **R**esins, **A**romatics oil analysis
FB	**F**ormation **B**ine
SW	**S**eawater
OSW	**O**ptimum **S**mart **W**ater
LSW	**L**ow **S**alinity **W**ater
3dSW	**3**-times **d**ilution of **S**eawater by distilled water
5dSW	**5**-times **d**ilution of **S**eawater by distilled water
3S-C-M	seawater with **3**-time the **S**ulfate concentration, **1**-times the **C**alcium concentration, and **1**-times the **M**agnesium concentration
3S-3C-M	seawater with **3**-time the **S**ulfate concentration, **3**-times the **C**alcium concentration, and **1**-times the **M**agnesium concentration
3S-C-3M	seawater with **3**-time the **S**ulfate concentration, **1**-times the **C**alcium concentration, and **3**-times the **M**agnesium concentration
6S-C-M	seawater with **6**-time the **S**ulfate concentration, **1**-times the **C**alcium concentration and **1**-times the **M**agnesium concentration
6S-C-3M	seawater with **6**-time the **S**ulfate concentration, **1**-times the **C**alcium concentration and **3**-times the **M**agnesium concentration
6S-3C-M	seawater with **6**-time the **S**ulfate concentration, **3**-times the **C**alcium concentration and **1**-times the **M**agnesium concentration
OOF	**O**ne **O**pen **F**ace
TOF	**T**wo **O**pen **F**ace
OOCO	**O**ne **O**pen face and another face of core isolated from brine with a special tube that allows collection of the **CO**-current production

1. Introduction

Complexities associated with the oil production mechanisms in majority of oil-wet carbonate reservoirs pose great challenges. The wettability alteration of oil-wet carbonate reservoirs toward water wetness would improve oil production when stickiness of oil to the rock surface decreases. In the past few years, smart water injection (modified water chemistry in terms of salinity and composition to prepare the most efficient brine composition for a specific oil/brine/rock system [1]) and low salinity water (low ions concentration) has attracted much attention from EOR (enhanced oil recovery) research community. There have been many investigations concentrating on wettability state of carbonate reservoirs [2–9] considering different methods to affect interfacial tension [10,11] and wettability state such as gas-based scenarios like miscible and immiscible gas injections into water or oil [12,13], or water-bases ones taking the advantages of different chemicals like surfactants and nanoparticles [14–16] or ions to change the reservoir condition towards oil production improvement. Since the brine modification is an available and affordable in most water-based scenario projects, most current researches are focused on figuring out new aspects of ions influence on the wettability state of reservoirs.

Wettability alteration during water-flooding by adjusting the composition of injected brine is becoming economically more viable thus has become a matter of much interest [17]. The main objective of EOR by ion exchange (chemical) mechanism is to improve the mobility of trapped oil during water-flooding [18–20]. In low permeability rock such as carbonate matrix as a result of using surfactant and brine, the interfacial tension (IFT) will be decreased (about 0.3-1.0 mN/m) and consequently the imbibition rate is expected to decrease even though the wettability condition changes to more water wetness [10,21,22]. By the use of diluted Persian Gulf seawater (SW) by 2, 10, and 20 times, the amount of oil recovery from composite limestone cores has been reported to increase [23]. Nonetheless, in a chalky cap rock flooded with diluted SW, the performance of flooding has been reduced and it did not have the same effect as what was to be for composite limestone [24,25]. Calcium and magnesium potentially active ions in the wettability alteration process which is required to get close to the rock surface for reaction with a carboxylic component of crude oil and form a metalorganic complex [19,26]. Although calcium and magnesium are thought to be ions which participate in wettability alteration (forming metalorganic complexes), it is assumed that there is also a possibility of sulfate contribution to the wettability alteration [27–29]. Due to the negative charge of the sulfate ions, they are able to get close to the carbonate surface and by alleviating the similar positive carbonate rock charge with calcium and magnesium, provide a suitable situation for easy reaction with carboxylic components [24,25,30]. Chemical activity and mobility of cationic ions strongly depend on the temperature of the bulk of fluids which are in contact with the rock surface [17,31].

Fractured carbonate reservoirs are composed of two porous media with different permeability conditions of i) fracture system with high permeability and ii) low permeable block matrix. Accordingly, during water injection, matrix blocks would be surrounded by water through the fractures and then two types of oil flow directions are expected to occur i) co-current and ii) counter-current oil flow. When water enters the carbonate block matrix, oil can be produced either in the same direction (co-current) or in the opposite direction (counter-current). The amount of oil production due to these mechanisms would depend on matrix size and permeability, oil/water viscosity ratio, type of ions that exist in the brine, crude oil chemistry, and reservoir pressure and temperature conditions that control the oil feed from matrix to fracture.

Many factors such as wettability [31–34], viscosity [35], IFT [33–36], oil composition, injection fluids properties are known to influence counter-current oil flow. In addition, the interaction between the fluid and porous media depends on pore shape, matrix permeability, relative permeability and boundary conditions, which would, in turn, determine the rate of imbibition and ultimate oil recovery [37]. At the beginning of the imbibition process, the counter-current oil flow always prevails. The oil/water mobility ratio determines how counter-current continues which otherwise depends on the viscosity of wetting phase. If brine mobility is high, then counter-current oil flow continues, otherwise it stops and instead co-current flow begins to produce oil [38,39]. Capillary back pressure (CBP) was mentioned by Parsons and Chaney [40,41] but it is usually ignored in co- and counter current imbibition due to the strength of water-wet condition and high-water saturation which does not exist in the early stage of imbibition. The surfaces where oil is produced as droplets the capillary back pressure (CBP) would occur at the open face. During the imbibition process large oil droplets would usually stick to the open face of the core. In this process, if the radius of curvature of oil droplets is much larger than the pore radius, then they would only produce small back pressures. Snap off phenomenon could also happen for strongly water-wet conditions near the faces that are in contact with the wetting phase, resulting in coalescence of large droplets on the outside of the core [42]. The study of imbibition has usually been in the form of all open faces core [43], therefore for a closer look, one open face core investigation is required although in this model the shape of recovery curve would depend on the core length to diameter aspect ratio [34]. The imbibition always starts counter-currently and its duration depends on fluids viscosity [28]. If water viscosity decreases though then all the interlayer fluid resistance will be in the oil and counter-current oil flow will continue until the oil/water front reaches the other end of the core [44]. Viscosity of the wetting phase is inversely related to co-current imbibition rate, in addition, the impact of pore geometry type and pore size core distribution is imperative for residual oil saturation in the porous medium [45].

Investigation the dominancy of either co- or counter currents under the influence of different water salinities having considered the effect of carbonate cores physical parameters is the subject that novelty of the present study relies in. In this study, after determination of optimum smart water composition and low salinity water concentrations, the static evaluation of wettability alteration (WA) by the use of Amott method and dynamic evaluation by relative permeability (Kr) was conducted. The impact of core face isolation on co- and counter-current oil flows was also investigated when optimum smart water and low salinity water imbibe into carbonate cores. Finally, the influence of core length and permeability on recovery factor was discerned in co- and counter-current oil flows when the optimum smart water imbibed into carbonate cores.

2- Methodology & Procedures

2-1- Stepwise road map

Figure 1 shows the followed procedures for clarifying the roles of the wetting phase which is imbibed by carbonate cores, as well as determination of core length and permeability effects on co-and counter-current oil flows.

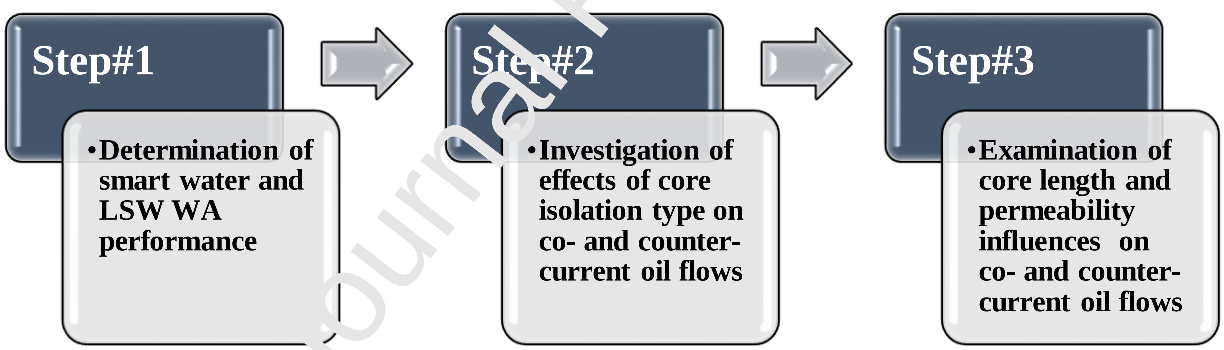

Figure 1: Stepwise road map for investigation the effects of brine type, core length and permeability influences on co- and counter-current oil flows.

2-2-Materials

2-2-1-Core samples

An outcrop block of Iran’s carbonate formation pertaining to early Oligocene geological period from which many plugs were taken has been investigated. The results of X-ray Diffractometry (XRD) test are shown in Table 1, which demonstrate that the sample contains up to 73% of

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calcite. The wafer-size thin sections (thickness of 1.5 mm) were cut for the measurement of contact angle, and their surfaces were polished for minimizing roughness effect [46].

The carbonate cores' petrophysical properties and fluids volume after flooding process (pore volume (V_p), formation water (V_w) and oil volume (V_o)) are provided in Table 2. In this study, the VINCI Keyphi apparatus was used for measurement of porosity and permeability and VINCI core flooding apparatus was used for saturating the cores with oil and brine.

Table 1: The used carbonate rock compounds percent obtained by XRD test

Compound name	Constituents percent
Calcite	73
Ankerite	[illegible]
Calcium Sulfate	1
Magnesium Zirconium Oxide	2
Lead Calcium Zirconium	3
Dolomite	19

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Table 2: Petrophysical properties of the used cores and volumetric oil and water saturation after flooding processes

Core No	Experiment	Φ (%)	K (mD)	L (mm)	V_p (cc)	V_w (cc)	V_o (cc)
1	Amott method	15.0	25	67	11.48	3.45	8.03
2	Kr Determination	15.0	28	67	11.48	0.80	10.67
3	Isolation behavior	17.4	32	67	13.28	2.60	10.68
4	Isolation behavior	18.3	31	67	13.97	2.70	11.27
5	Isolation behavior	19.0	33	67	14.50	2.90	11.60
6	Isolation behavior	16.9	29	67	12.90	2.60	10.30
7	Isolation behavior	17.9	32	67	13.66	2.70	10.96
8	Isolation behavior	18.0	30	67	13.74	2.80	10.94
9	Co- & counter flow	21.0	29	50	13.20	2.40	10.80
10	Co- & counter flow	23.0	33	70	19.00	4.40	14.60
11	Co- & counter flow	24.0	30	90	24.62	4.18	20.44
12	Co- & counter flow	22.0	72	70	17.55	2.60	14.90

2-2-2-Fluids properties

2-2-2-1- Brine

As stated earlier, Calcium and magnesium are potentially active ions in the wettability alteration, it is assumed that there is also a possibility of sulfate contribution to the wettability alteration [17,27–29]. Based on this supposed theory, the design of smart water has been done in such a way that the roles of magnesium, calcium, and sulfate in the wettability alteration process are examined. To investigate and compare the performance of smart water and LSW:

1) Representative of LSW samples was selected among the seawater (SW), 3 times (3dSW) and 5 times (5dSW) of diluted seawater (Table 3). These combinations are intentionally being selected since we know that oil recovery decreases by increasing dilution [47]. Furthermore, for better comparison between smart water and diluted SW performances and to determine the importance of active ions [19] concentration, the dilution more than 5 times was not selected.
2) For investigation of divalent ions effect, smart water representative was selected between seawater with 3-time the sulfate concentration, 1-times the calcium concentration and 1-times the magnesium concentration (3S-C-M), and in the same way (3S-3C-M), (3S-C-3M), 6-time the sulfate concentration, 1-times calcium concentration and 1-times magnesium concentration (6S-C-M), and in the same way (6S-C-3M), (6S-3C-M). All ionic strength were kept constant equal to that of seawater (0.63 $mol.L^{-1}$) by adjusting the concentration of NaCl. Table 4 represents the ions concentrations of each designed smart water samples.

The initial water saturation in the cores was also obtained using formation water of one of the southern Iranian fractured carbonate oil fields with ionic strength of 2.8 $mol.L^{-1}$. The concentrations of various investigated brines are given in Table 3 and Table 4.

Table 3: Concentration of various diluted brines

Composition	SW	3dSW	5dSW
Na^+ ($g.L^{-1}$)	17.00	5.67	3.40
Ca^{2+} ($g.L^{-1}$)	0.55	0.18	0.11

Mg^{2+} (g.L^{-1})	1.81	0.60	0.36
K^{+} (g.L^{-1})	0.43	0.14	0.09
SO_4^{2-} (g.L^{-1})	3.20	1.07	0.64
HCO_3^{-} (g.L^{-1})	0.75	0.25	0.15
Cl^{-} (g.L^{-1})	23.25	7.75	4.65
Ionic strength (mol. L^{-1})	0.63	0.21	0.12

Composition	6S-C-M	6S-3C-M	6S-C-3M	3S-C-M	3S-3C-M	3S-C-3M
Na^{+} (g.L^{-1})	14.43	12.13	10.76	16.52	15.36	12.25
Ca^{2+} (g.L^{-1})	0.55	1.59	0.55	0.55	1.59	0.55
Mg^{2+} (g.L^{-1})	1.81	1.81	5.43	1.81	1.81	5.43
K^{+} (g.L^{-1})	0.43	0.43	0.43	0.43	0.43	0.43
SO_4^{2-} (g.L^{-1})	19.85	19.85	19.85	9.93	9.93	9.93
HCO_3^{-} (g.L^{-1})	0.75	0.75	0.75	0.75	0.75	0.75
Cl^{-} (g.L^{-1})	21.95	25.3[illegible]	26.89	20.91	21.14	22.94
Ionic strength (mol. L^{-1})	0.63	0.63	0.63	0.63	0.63	0.63

Table 4: Concentrations of designed smart water samples

2-2-2-2- Crude oil

The used crude oil was obtained from one of the southern Iranian fractured carbonate oil fields with °API and viscosity values of 23.7 and 96 cP at ambient condition, respectively. The saturates, asphaltenes, resins, aromatics (SARA) [48] oil analysis showed that the amount of asphaltene was about 7%.

2-3-Procedures

Following are the pre-required and designed procedures to examine the effects of different parameters on co- and counter-current oil flows. The schematics of all followed procedures are demonstrated in Appendix section from Figure to Figure 18.

2-3-1- Aging process

I. Formation brine-saturated carbonate cores and thin sections were maintained initially for 14 days within a beaker full of formation brine (FB) at 75°C to achieve their original state like what were before migration process [17,24,29].

II. The FB in the cores was replaced by the crude oil using core flooding system (oil migration process). Having cores pore volume and volume of injected oil, the amount of irreducible FB saturations and injected crude oil have been calculated (Table 2).

III. Next step after oil flooding, oil-saturated cores and thin sections were then kept in crude oil at 75°C and 2500 psi for 20 days. It should be pointed out that the amount of oil-wetness changed due to the different composition of each thin section.

IV. After the accomplishment of the aging process, due to the reduction of those crude oil components which were adsorbed to the carbonate rock surface and also considering that fresh oil would exist in the reservoir, the oil that was injected into the cores for aging process, was replaced with the fresh oil.

2-3-2-Procedures of Step1: Potential Evaluation of Smart Water and LSW for WA

2-3-2-1- Contact Angle Measurement

The Contact angle (CA) measurement procedure was carefully performed to discern the optimum concentration of various designed brines (smart water and LSW) for maximum wettability alteration:

i. Water from the nearest and the most accessible sea (Persian Gulf) with known ions was taken as a sample.

ii. In LSW samples, dilution for 3 and 5 times were performed using distilled water (Table 3).

iii. In smart water samples (Table 4), all brines were synthetic. By adjusting the salt concentration, ionic strength (the total number of moles of salt per liter) were kept constant with decreasing or increasing concentrations of sodium chloride.
iv. All smart water samples were of the same ion strength with different concentration ratios of sulfate, calcium, and magnesium to determine the best interaction with carboxylic acid groups for removing them from carbonate thin sections.
v. All thin sections in the specified time interval of 0, 48, 120, 360 hrs were placed in the designed brines before measuring the contact angle.

In this study, a drop shape apparatus (DSA100, KRUSS, Germany) was used for the measurement of contact angles. In brief, for contact angle measurement, a micro-syringe was fitted with a U-shape needle and loaded with the fluid with lower density (i.e., crude oil). The syringe was then placed in a motor-driven piston and the tip of the U-shaped needle was positioned in an optically clear vessel and immersed in the aqueous phase. A carbonate rock thin section with the thickness of about 1.5 mm was then placed on a rubber block before continuing brine injection to the point at which the drop is big enough and leaves the needle sticking to the thin section surface. Finally, the image of the drop on the thin section was analyzed. After finding the best salinity of active ions such as sulfate, calcium and magnesium for WA and IFT reduction by contact angle and pendant drop method, to check the accuracy of the resultant findings, the Amott method and relative permeability test were also conducted.

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2-3-2-2- Amott method

To check the performance of determined optimum brine using contact angle model in pore scale at static condition the Amott wettability index (AWI) was calculated as follow:

Oil-wet core of #1 (number 1) was soaked for 15 days in the optimum smart water (3S-C-3M) before performing the following processes in sequence [49]:

I. Oil flooding to calculate the amount of irreducible optimum brine within the core after forced drainage.
II. Imbibition cell to obtain the amount of distilled water spontaneous imbibition.
III. Water-flooding to obtain the volume of distilled water forced imbibition.

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IV. Drainage cell to obtain the volume of oil spontaneous drainage.

Distilled water was used because it had no effect on wettability during the test. Table 5 shows the wettability condition determination range by Amott index [49].

Table 5: AWI conditions [49]

AWI Ranges	Wettability state
0.3 to 1.0	Water-wet
-0.3 to 0.3	Neutral
-1 to -0.3	Oil-wet

2-3-2-3- Relative permeability test

The relative permeability [50] test is extensively used at dynamic condition for determination of wettability alteration. To examine the effect of obtained optimum brine in pore scale at dynamic condition, the core #2:

i. At first, was flooded with formation water (FW) and at each time step by fraction fluid collector the volumes of the oil and water production were calculated and also using pressure gauge apparatus related pressure data were recorded, simultaneously.

ii. In the second step, the core was saturated with optimum smart water (3S-C-3M) and soaked for 15 days and in the third step the core was flooded with oil and the first step was repeated.

iii. Thereafter, the resultant intersections between oil and water relative permeability curves in the first and third steps were compared.

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2-3-3-Procedures of Step2: Investigation of core faces isolation type on co- and counter-current oil flows

In this section, to investigate the impact of determined representatives of designed smart water samples and LSW on the amount of co- and counter-current produced oils, cores # 3, 4, and 5 for optimum designed smart water imbibition and cores # 6, 7, and 8 for optimum LSW imbibition were coated with different faces isolation type as follow:

I. Lateral surface along with one face of cores# 3, and 6 were coated using epoxy resin→OOF
II. Lateral surface of cores# 4, and 7 were coated using epoxy resin so that two core faces allowed to be in touch with brine→ TOF
III. One open face and another face of cores# 5, and 8 isolated from brine with a special tube that allows collection of the co-current production→OOCO

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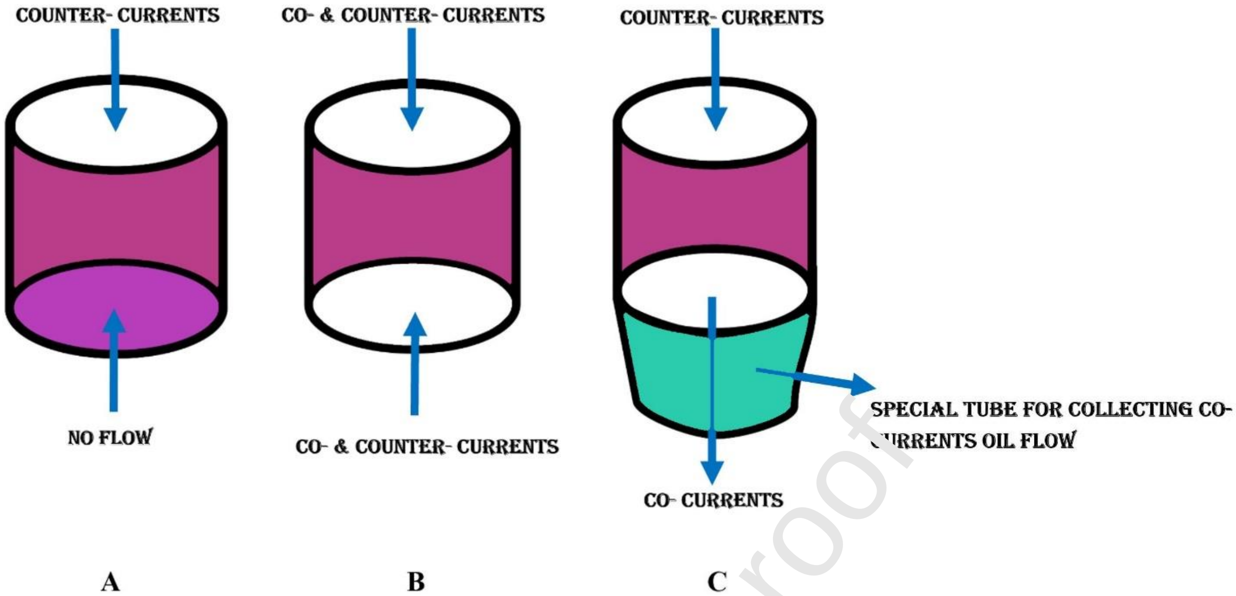

Figure 2: Red color shows the lateral surfaces and faces that coated with epoxy resin. A) one open face (OOF) B) two open faces (TOF) C) one open face and another face isolated from brine with a special tube (green color) that allows collection of the co-current production (OOCO)

2-3-4-Procedures of Step3: Examination of core length and permeability influences on co- and counter-current oil flows

The imbibition tests are usually performed in all open faces and in such experiments, the amount of oil production by co- and counter-currents are not determinable individually. After determination of the best wetting phase type between diluted seawater and smart water samples from the step1, the next step was to investigate the effect of core length and permeability on co- and counter-current individually. Cores #9, 10, and 11 were used for core length investigation which possessed similar permeability, and also the comparison between the results of oil production for equal length cores #10, and 12 showed the permeability change effect. For this purpose:

i. Boundary conditions were considered in such a way that core lateral surface was coated with epoxy resin and tube and one face was kept open in contact with the brine whereas the other with the crude oil inside an oil filled plastic container.

ii. Two inverse graded funnels were used to collect the co- and counter oil production separately, as shown in Figure 3.

iii. The prepared core was then submerged in the optimum smart water (3S-C-3M) to initiate co- and counter-current flows of oil. The face that is contacted with brine produce the oil counter-currently and the other face has co-current oil production.

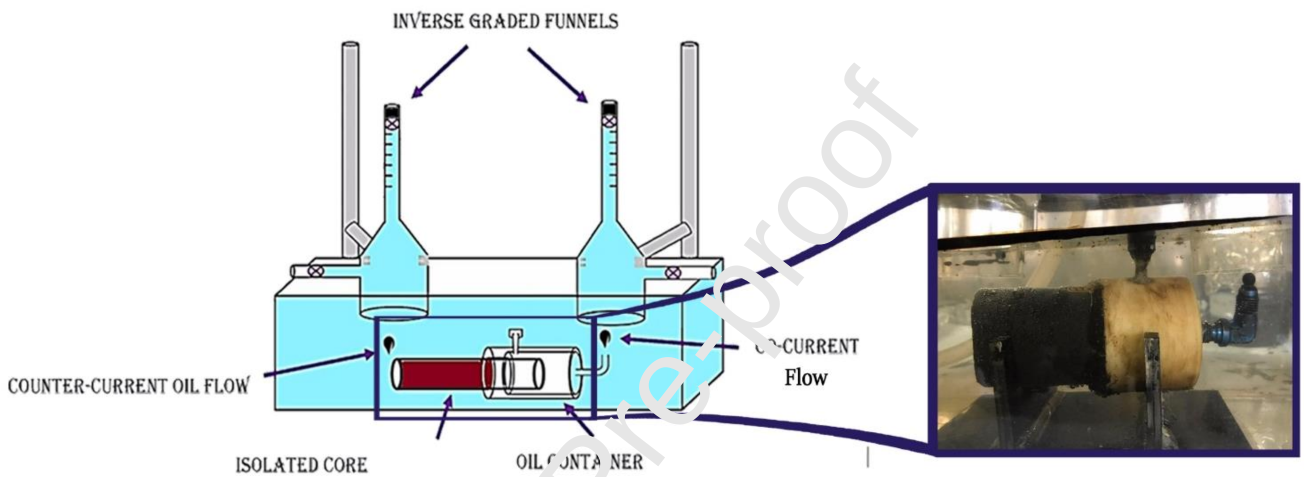

Figure 3: Co- and counter current imbibition setup

3-Results and Discussion

3-1-Results of Step1: Potential Evaluation of Smart Water and LSW for WA

3-1-1-WA by LSW

Figure 4 shows the WA for SW and 5dSW (5-times diluted seawater) and 3dSW (3-time diluted seawater). The superiority of 5dSW in WA than SW and 3dSW is probably due to the concentration of ions near the rock surface and more chance for active ions like magnesium, and calcium to reach to the carboxylic components which were bound to the carbonate rock and separate them from carbonate rock [36] and alter the wettability from oil-wet to water-wet .

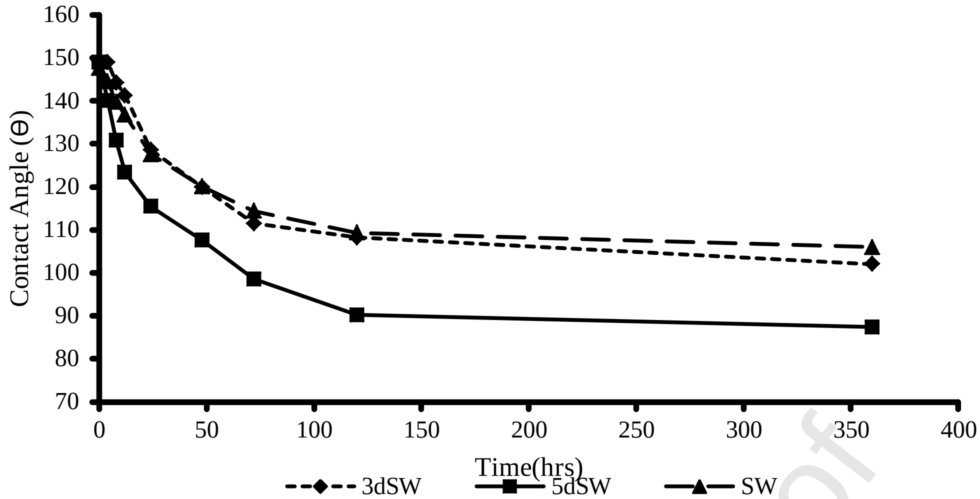

Figure 4: Wettability alteration of sea water and diluted seawater samples

3-1-2-WA by Smart Water Samples

Different concentration ratios of active ions (Table 4) for similar ion strength showed the importance of the type of divalent cations for intensifying wettability alteration. As it can be seen from Figure 5, 3S-C-3M has the maximum wettability alteration from 129 to 30 degrees.

As shown in Figure 5, in comparison with 6-time sulfate samples, wettability alteration for magnesium due to its chemical activity and surface density charge [51] for 6S-C-3M is higher than for 6S-3C-M. For all synthetic brines, magnesium with compared to calcium played a more important role in WA.

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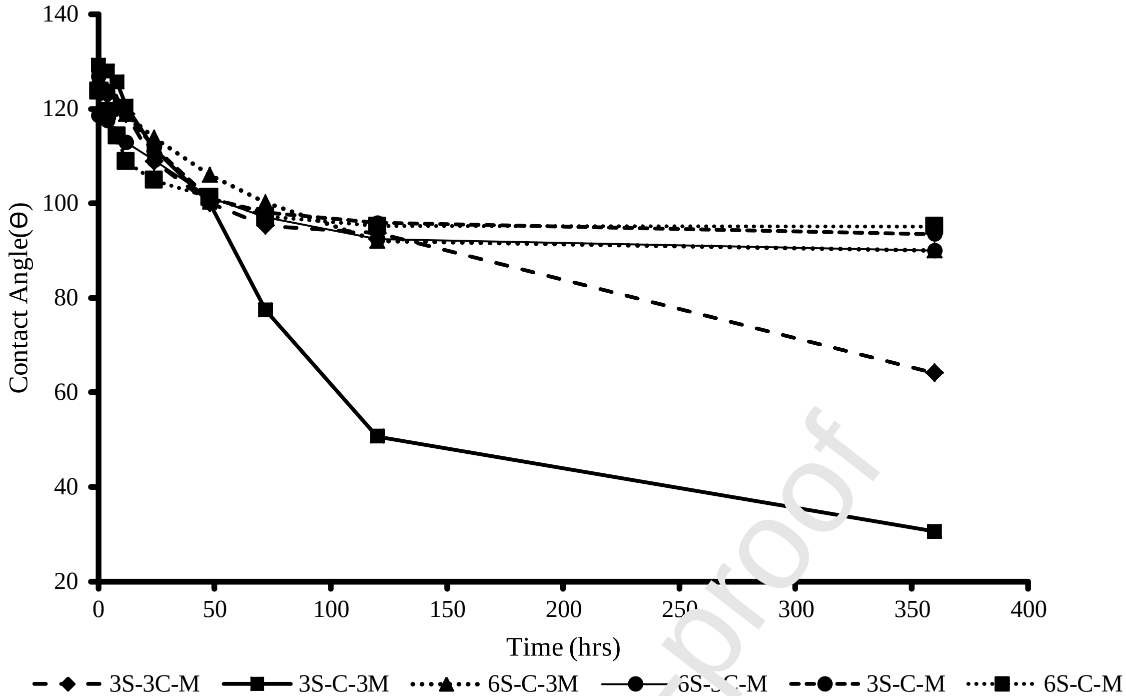

Figure 5: Wettability alteration of (3S-C-M), (3S-3C-M), (3S-C-3M), (6S-C-M), (6S-C-3M), (6S-3C-M)

3-1-3-Wettability Alteration Index

Due to mineral heterogeneity of the thin sections which, not surprisingly, resulted in different compositions, then the contact angles (oil-wet condition) after ageing process will be different. As a result, the difference between aged and final (water-wet condition) angles cannot serve as a reliable indicator for comparison of WA. Therefore, a non-dimensional number is defined as Wettability Alteration Index (WAI), which is defined as the difference between aged and final angles divided by the difference between aged and initial (before aging process) angles. Figure 6 shows the resultant WAI for the diluted seawater and smart water samples. As illustrated in this figure, 3S-C-3M and 5dSW with WAI values equal to 0.82 and 0.45 are the best representatives for smart water and diluted seawater, respectively. The comparison of SW ("S-C-M"→WAI=0.3) and 3-time sulfate ("3S-C-M"→WAI=0.44) and 6-time sulfate ("6S-C-M"→WAI=0.38) implies that sulfate anions would highly tend to get closer to the carbonate surfaces [36]. Thus, the sulfate anions must be at a proper concentration for permitting the accessibility of other ions.

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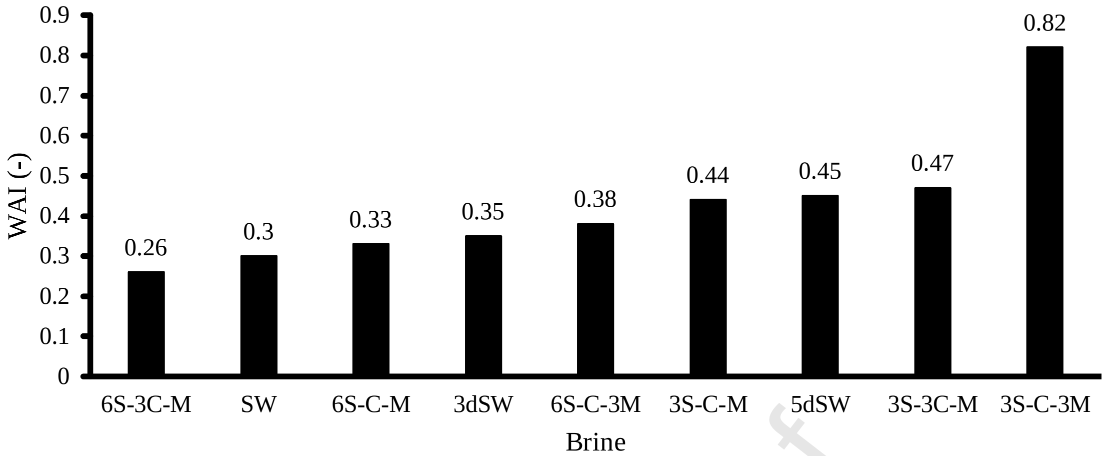

Figure 6: Wettability alteration index of the designed samples

3-1-4-IFT Results

Measuring IFT values could be measured during the time that its value is changing. However, in this study the final IFT in each case has been measured and reported at the time that it was ensured there is no IFT variation anymore [52].

3-1-4-1-IFT of diluted sea water sample/ oil systems

The maximum IFT among the diluted seawater samples belongs to 5dSW (see Figure). The appropriate IFT for an imbibition process is in higher IFT value (increasing IFT (б) strengthens the capillary pressure ($P_c = {2\sigma \cos\theta}/{r}$, $CA = \theta$, $\text{Pore radius} = r$) and cause the tendency intensifying of pores to imbibe the wetting phase) and the WAI for 5dSW sample has the maximum value as well. Therefore, the 5dSW was considered as an optimum representative of diluted samples (see Figure).

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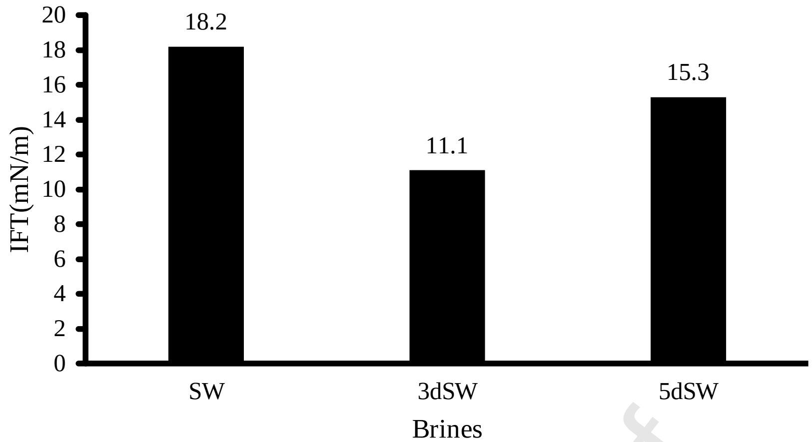

Figure 7: IFT of the designed LSW samples/oil system

3-1-4-2- IFT of smart water sample/oil systems

The IFT results for smart water samples are consistent with those of WAI results. 3S-C-3M presents the minimum IFT value (8.3 mN/m) and maximum WAI among all samples. Therefore, 3S-C-3M sample was considered as an optimum representative among the designed smart water samples. Figure , and Figure show the IFT the smart water sample/oil systems and the wettability and IFT results of the optimum representative (i.e. 3S-C-3M) with the oil sample, respectively.

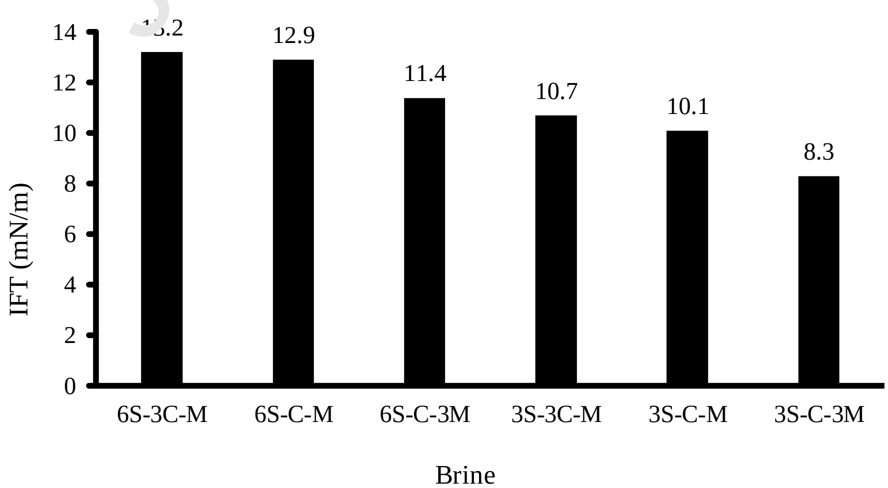

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Figure 8: IFT of the designed smart water samples/oil system

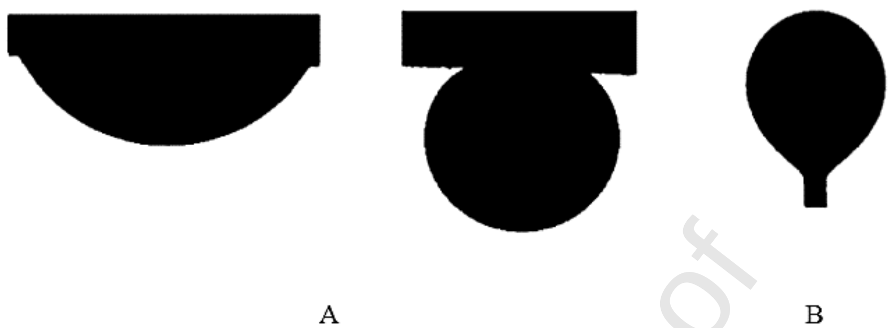

Figure 9: 3S-C-3M sample A) change of contact angle in a calcite thin section (from 129 to 30) B) pendant drop IFT measurement (at 3 mN/m)

3-1-5- Results of Amott test

During the Amott test, the measured fluids volume in forced drainage, spontaneous imbibition, forced imbibition and spontaneous drainage processes on soaked core in optimum smart water representative (i.e., 3S-C-3M) led to values of 7.6 cc, 3.86 cc, 2.85 cc, and 0.8 cc, respectively. More spontaneous imbibition than spontaneous drainage yields Amott index of 0.37, which confirm the water-wetness wettability state of core sample.

3-1-6- Relative Permeability Test

As can be observed from Figure 7, changing the intersection of the water and oil relative permeability curves from 0.46 to 0.54 on the water saturation axis shows the positive effect of core soaking in smart water, which resulted in wettability alteration towards water-wet conditions.

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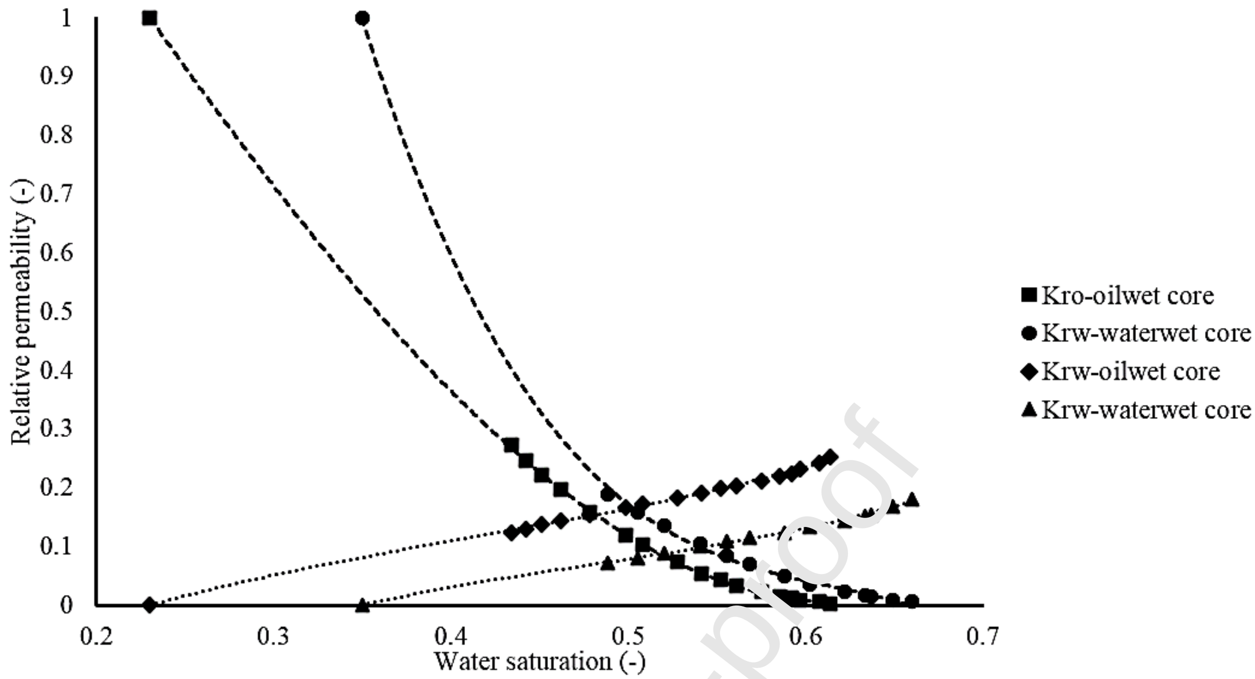

Figure 7: Water and oil relative permeability of water-wet and oil-wet core

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3-2-Results of Step2: Effects of core faces isolation type on co- and counter-current oil flows

3-2-1- Investigation of co- and counter-current flows on each other

Two groups of cores were submerged in the Amott cell. The cells were filled with the synthetic brines to the same level to eliminate gravity effects and were kept at a constant temperature of 75°C. Then, for about 12 days, the amount of produced oil was recorded.

For all brines that were used in these experiments, their viscosity was more or less equal to distilled water due to the low level of total dissolved solids. Thus, all resistances against entrance of the brines into the cores were in the oil phase [42], and all flow was due to capillary pressure gradient. Due to further wettability alteration in 3S-C-3M sample, it was expected for the capillary pressure gradient to be greater than that in 5dSW imbibition.

More oil recovery in TOF cores could be due to the existence of co-current and counter-current oil flows in two faces simultaneously [53] (Figure 8-A, and B). By entrance prevention of brine from one face (OOF), co-current oil flow would be eliminated and hence oil will be produced by counter-current oil flow. On the contrary, when the other face in a special condition without contact with brine was allowed to produce co-current oil, the amount of counter-current oil production was reduced on the opposite face of the core.

The reason could be that when co-current oil flow was allowed (OOCO) to the cores, then the imbibition begins to prevail due to the impact of oil viscosity [42], the counter-current oil flow will start [42]. As time elapses, though, the smart water invades the core due to increased water-wetness ,based on that , capillary pressure gradient will strengthen and will be able to overcome oil interlayer friction [25], which exists after the oil/water front and consequently oil would be produced co-currently.

After smart water penetrates through the core, the amount of resistance for the co-current oil production is less than the resistance against oil counter-current production. This is because after a while of counter-current oil production, fluid fronts move forward through the cores thus i) the oil volume that have to be replace, which is in the pores after the oil / water front, decreases and ii) capillary pressure would not have enough force to overcome the resistance of water and residual oil droplets interfaces behind oil/water front. The movement of oil-water front in imbibition process is piston like [54] though oil droplet upward movement would weaken due to parameters such as trapping oil mechanisms, interlayer friction and mixed wettability conditions. However, the buoyancy force and capillary pressure are those which facilitate oil production.

An important issue is the competition between the oil interlayer frictions as an obstacle against the oil flow and capillary pressure gradient as a booster in progress of imbibition process. During counter-current oil production, as the process goes forth and the front descends in the core, interlayer friction of the oil volume increases. This is because of a longer path ahead of the oil droplets than before while capillary pressure increases due to more water-wetness condition behind the front. Nevertheless, increase in these two opposing factors is unequal and capillary pressure gradient which imbibes the brine and ejects the oil out of the core seems not to be high enough.

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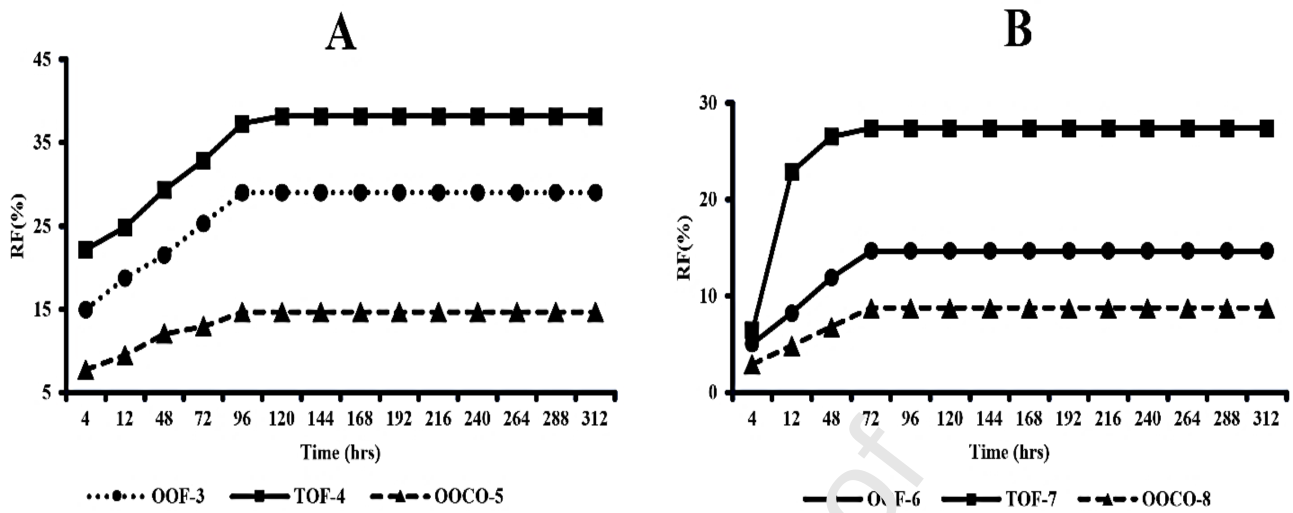

Figure 8: RF versus time for:

A) Smart water recovery curve of (Isolation type – core number) OOF-3, TOF-4, OOCO-5

B) 5dSW recovery curve of (Isolation type – Core NO) OOF-6, TOF-7, OOCO-8

3-2-2- Checking the Cross-sectional Area of Cores after Termination of the Imbibition Process

After accomplishment of imbibition process for the cases of TOF and OOCO, the two cores which were imbibed by optimum smart water were with specific distances of 1cm, 3.5cm, and 6cm from each core face. As Figure 12 depicts, in the core with TOF condition, the cross sections which are close to the faces (Figure 12-I, and III) more area has been washed by imbibed optimum smart water. Furthermore, the scrutiny of the cross-sectional areas indicates that those segments that their compaction during sedimentation was more intense, either from starting off the imbibition process were saturated with irreducible water or during the process their wettability states has been altered towards water-wetness and have been caused the oil discharge from larger pores. Figure 12-II outlines the fact that more distance from the faces causes less region affected by imbibed water than those of Figure 12-I, and III.

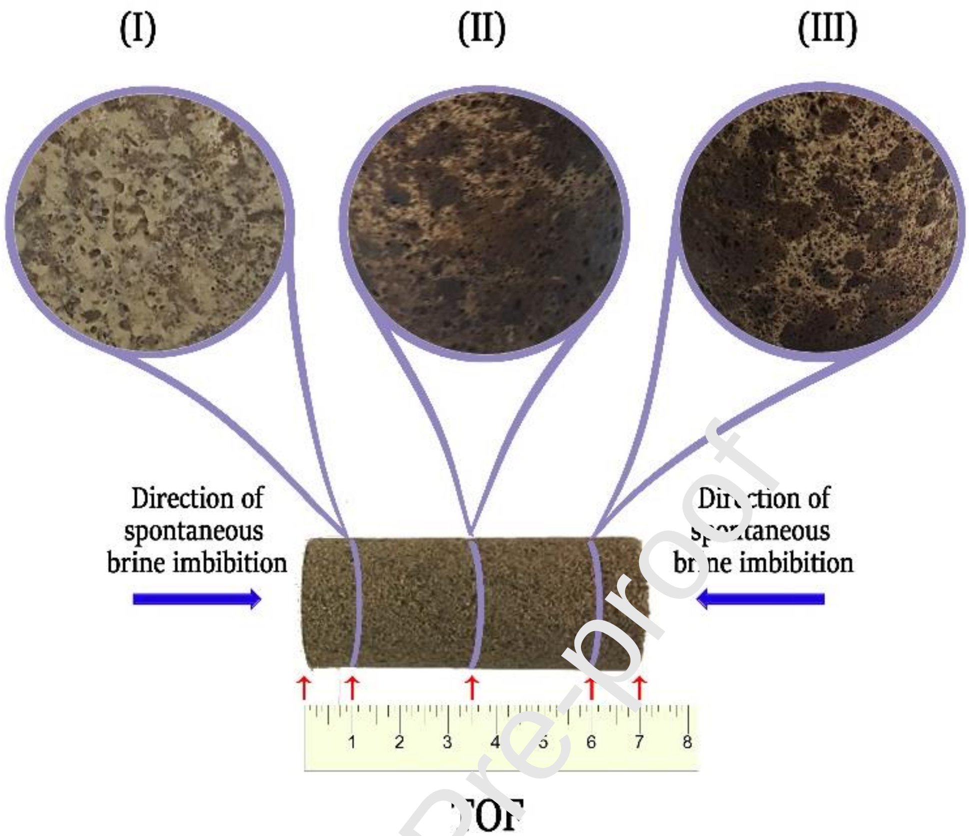

Figure 9: The cross-sectional areas with a distance of 1cm, 3.5cm, and 6cm from each core face with TOF

As it can be observed from Figure 13, in case of OOCO core isolation, the invading of imbibed optimum smart water has been weakened due to the removal contact of one face with brine. Distinguishable difference of washed area can be seen from Figure 13- I, and III. The cross section that has the shorter distance with the source of the imbibed optimum smart water (Figure 13- I) has been affected by the entrance of the brine while Figure 13- II, and III were the conduit of oil co-current flow. Generally speaking, the role of small pores along with the distance from the face that is connected to the source of the imbibed phase is the determinant of how the oil in the core will be swept by brine.

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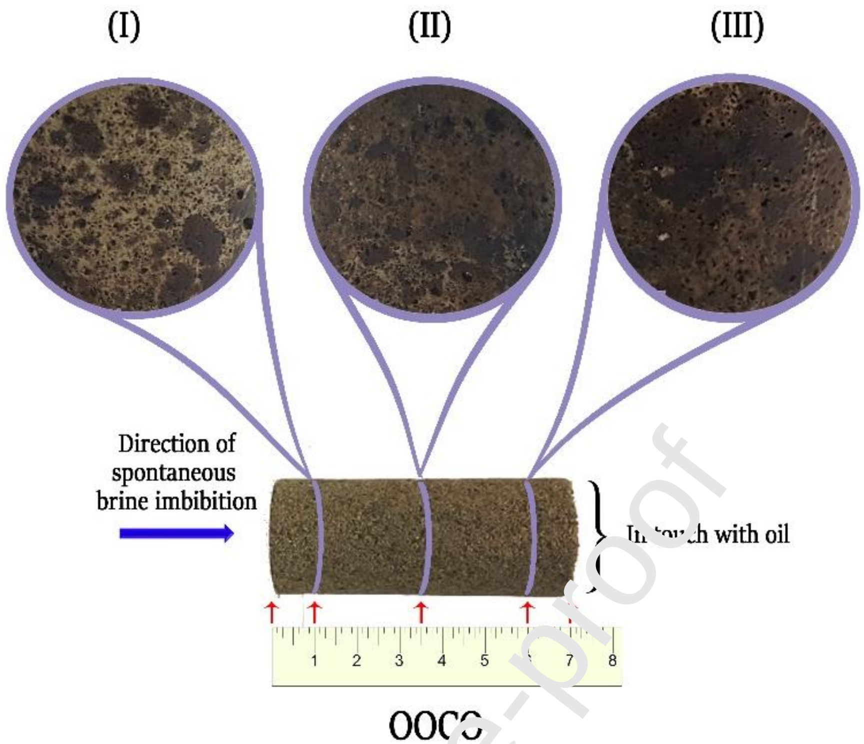

Figure 10: The cross-sectional areas with a distance of 1cm, 3.5cm, and 6cm from each core face with OOCO

With inspiration of the imbibition results, the schematic of co- and counter- current oil flows have been shown in Figure 14.

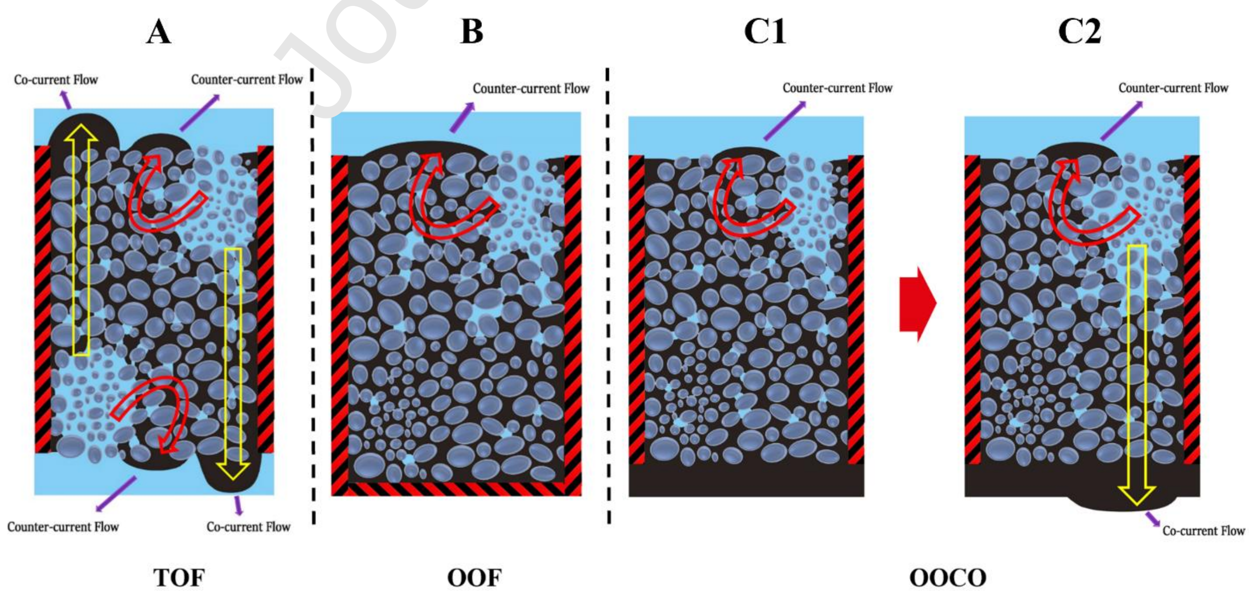

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Figure 11: Effect of different isolation core face on the amount of co- and counter oil flows A) TOF, B) OOF, C1) OOCO-earlier time of imbibition, C2) OOCO-after a while of imbibition

3-3-Results of Step3: Examination of core length and permeability influences on co- and counter-current oil flows

3-3-1-Impact of cores length

The experimental results using cores #9, 10, and 11 showed that with decrease of the core length, the amount of counter-current oil production will be decreased so that in the 5 cm core the counter-current flow reached approximately zero and co-current recovery factor intensified by as much as 20% and 13% more than in cores #10 and 11, respectively. By increasing the core length, with one face water imbibition, the amount of oil in place that has to evacuate from the other face will be increased. This means that for co-current oil production, the invading water should overcome the interlayer friction of the oil that exists between the oil/water front and the opposite core face which can be attributed to increased oil volume along the core. The required energy for movement of the wetting phase into core will be provided by capillary pressure gradient [56]. If the amount of capillary pressure gradient along the cores is insufficient to overcome the oil interlayer friction, then the co-current producing oil will not happen and oil will be forced to counter-current producing oil from the larger pores that exist in the same face which water imbibes into core. Nonetheless, CBP may weaken the counter-current flow at same larger pores. Comparison of initiation times of co-current oil flows in Figure 12-A, B, and C indicate that, the core#11 had a delay to start co-currently oil production due to the more resistance along the core in the oil phase. As it can be observed in Figure 12-C, after a while the beginning of the counter-current oil flow, the co-current oil flow also is started. This means that, after invading optimum smart water into the core and strengthening the capillary pressure gradient, the amount of oil and consequently its resistance to co-current flow has been reduced. Therefore, the co-and counter-current oil production has been observed simultaneously.

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Water imbibition by high capillary forces of the small pores and also replacing the water in the pores that evacuate during counter-current producing oil are two reasons for increased water saturation. As a result, the water-wetness would intensify and more oil will be produced co-currently. Figure 12-A, B, and C present the co- and counter-current oil flows for cores # 9, 10 and 11.

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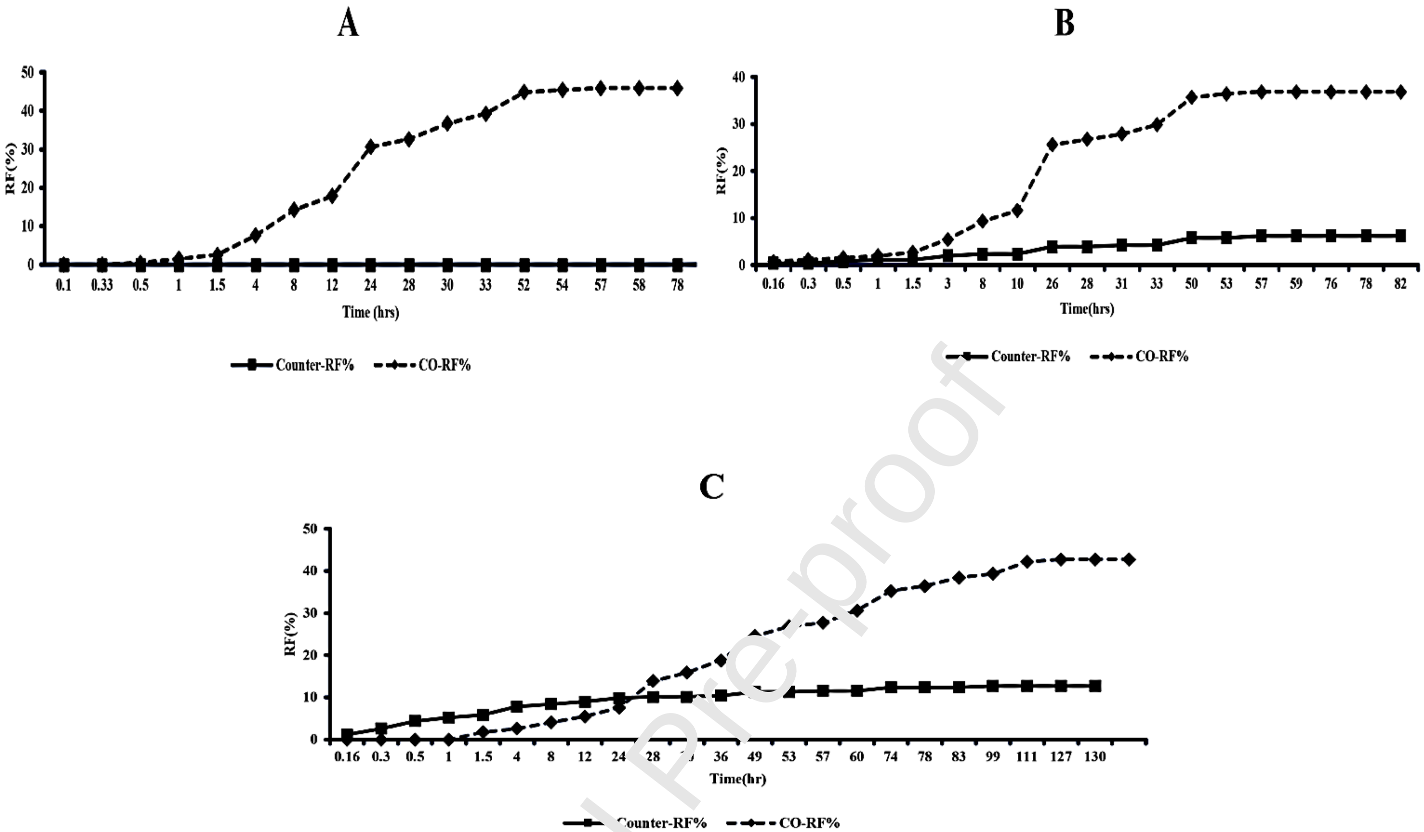

Figure 12: RF vs. time of co- and counter-current oil flows curves for (Core Number-Length-Permeability)

A) core #9- Length=5cm-Permeability=29mD

B) core #10 - Length=7cm-Permeability=33mD

C) core #11- Length=9cm-Permeability=30mD

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3-3-2-Impact of core permeability

Figure 13 compares the oil recovery of cores # 10 and 12. The length of these two cores was similar with different permeability. As it can be observed from this figure, the share of the counter-current oil production increased sharply by 8% in core #12 with more permeability. It is

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well known that, as permeability increases, the diameter of the pores in the core increases [56–58] and as a result the capillary pressure gradient will be weakened hence the ability of the capillary force for co-current oil production reduces.

Table 6 compares the share of co- and counter-current recovery factor from the total oil production (TP) and from the original oil in cores (OOIC) at the beginning of the test. Comparative effects of core length and permeability variation on co- and counter-current oil flows are schematically shown in Figure 14.

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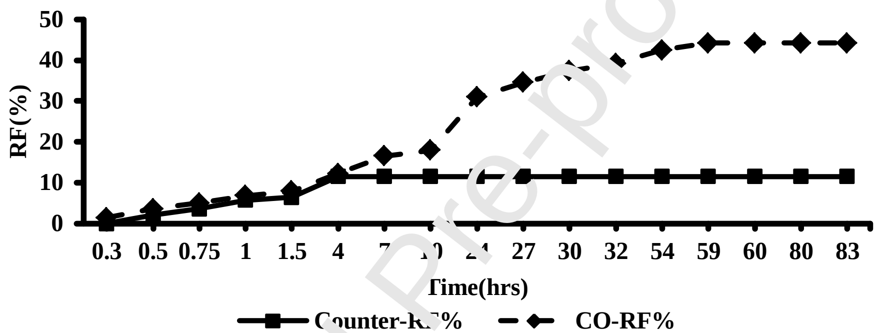

Figure 13: RF vs. time of co- and counter-current oil flows for core #12- Length=7cm-Permeability=72mD

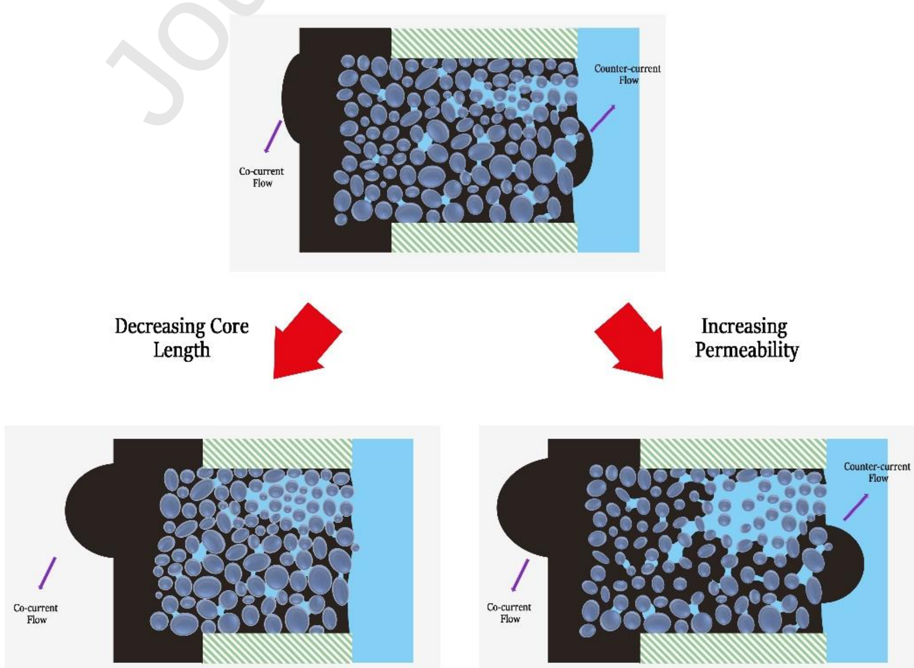

Figure 14: Effects of petrophysical properties changes on co- and counter current oil flows

Table 6: The shares of co- and counter-current recovery factor (RF)

Core #NO	$\%RF_{\text{TP (Counter-current Share)}}$	$\%RF_{\text{TP (Co-current Share)}}$	$\%RF_{\text{OOIP (Counter-current Share)}}$	$\%RF_{\text{OOIP (Co current Share)}}$
9	0	100	0	45
10	13	87	6.2	36
11	23	77	12	42
12	21	79	11.5	44

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Physical properties of a matrix, especially the part of its volume which is taken up by small pores is able to control the dominance of co-current or counter current oil production. Existence of a well-connected network of small pores and its location inside the matrix and how much it is possible to change the wettability state using different chemicals, gives the fractured reservoirs the capabilities to eject more volume of the available oil in the pores. Generally speaking, the closer small pores to the matrix face, the more counter-current oil production from that face, however, in addition to the advantages of small pores network, the other effective items that help the reserved oil to be produced are the average matrix permeability and wettability state as well in which having higher permeable matrix, will increase the expectation of a dominant co-current oil production. The competition between viscous forces (related to internal viscosity of oil) and capillary forces, distance of small pores from matrix faces, absolute permeability and the composition of the injected water could significantly affect the oil production from carbonate reservoir so that having more absolute permeability, reasonable amount of well-connected small pores and a modified brine according to the matrix composition compatibility , are the best conditions that could happen altogether to have a better oil evacuation of fractured carbonate reservoir.

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4- Conclusions

Matrix length, composition, and permeability are the key parameters that have to be taken in to account in order to finding suitable injection points in fractured reservoirs. Moreover, taking the advantages of mentioned ions in this study is the other side of the successful water flooding plans so that enables us to fully utilize the water injection operations with less amount of residual oil.

Counter-current and co-current flows depend on the chemical properties (brine composition and its effects on the wettability state and oil / water IFT) and physical properties (permeability of porous media, dimensions of block matrix) which govern in the filed system. Thus, based on the obtained results in the present study, in the filed scale, counter and co currents will exist and each one has a significant share in the total oil recovery. Therefore, since changing the physical characteristics of the reservoir is not economical and, in many cases really impossible, the importance of purposeful design of smart water that is selected to inject into the oil fractured reservoir will raise for the strengthening of the forces that play a significant role in the production mechanisms. To recapitulate, based on the experimental results of this study, the following conclusions can be drawn:

- Generally speaking, for smart water compared to diluted seawater, the total recovery with the use of optimum ions increased as a result of wettability alteration and IFT reduction.

- The presence of an optimal concentration of sulfate ions seems to be crucial for magnesium and calcium to increase the value of WA towards water-wetness.

- When lateral surface of cores was coated and:

  i) One face was in contact with brine and the other face was isolated (OOC) then only counter-current took place.

  ii) Two faces were in contact with the brine (TOF), at two faces co-current and counter-current happened.

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iii) One open face was in contact with brine (counter-current oil flow) and another face isolated from brine with a special tube (OOCO) that allowed collection of the co-current produced oil during the imbibition test, the co-current oil flow weakened the counter-current oil flow from the side that was in contact with the brine.

- The core length controlled the counter-current flow where the counter-current flow reduced by shorter core lengths. The results also showed that for core lengths below a critical value then the counter-current oil flow could utterly be eliminated.

- Core permeability influenced the imbibition process in such a way that by increasing the permeability the capillary pressure decreased resulting in intensified counter-current flow.

Declaration of competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix

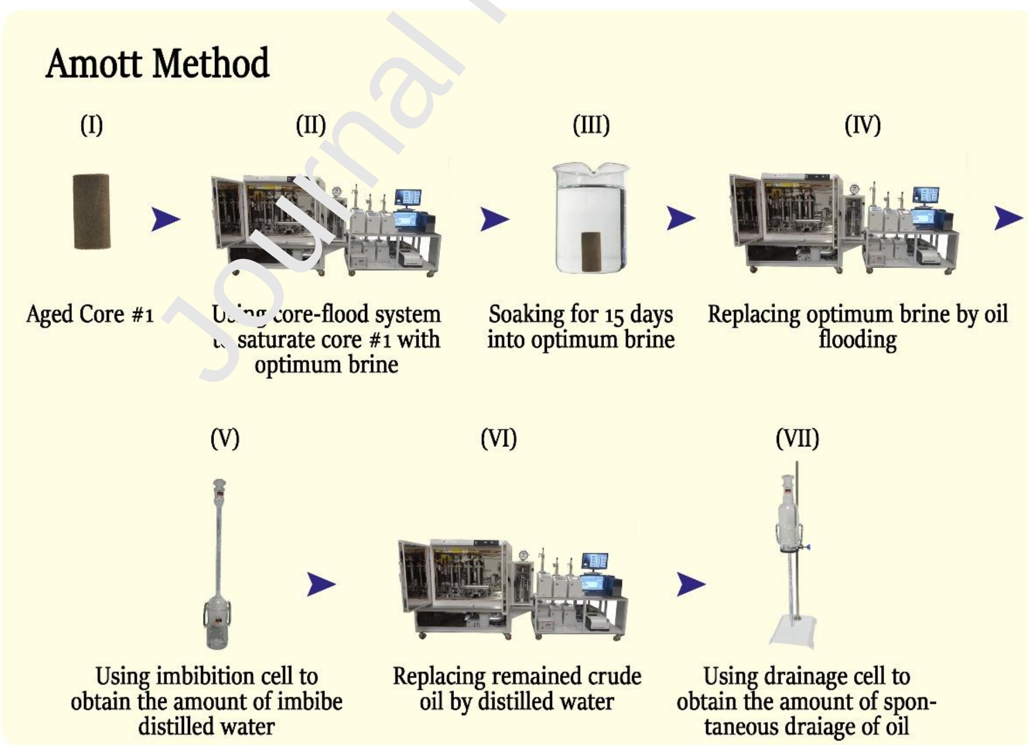

Figure 18: Steps of the aging process to reach the original condition after oil migration in a reservoir

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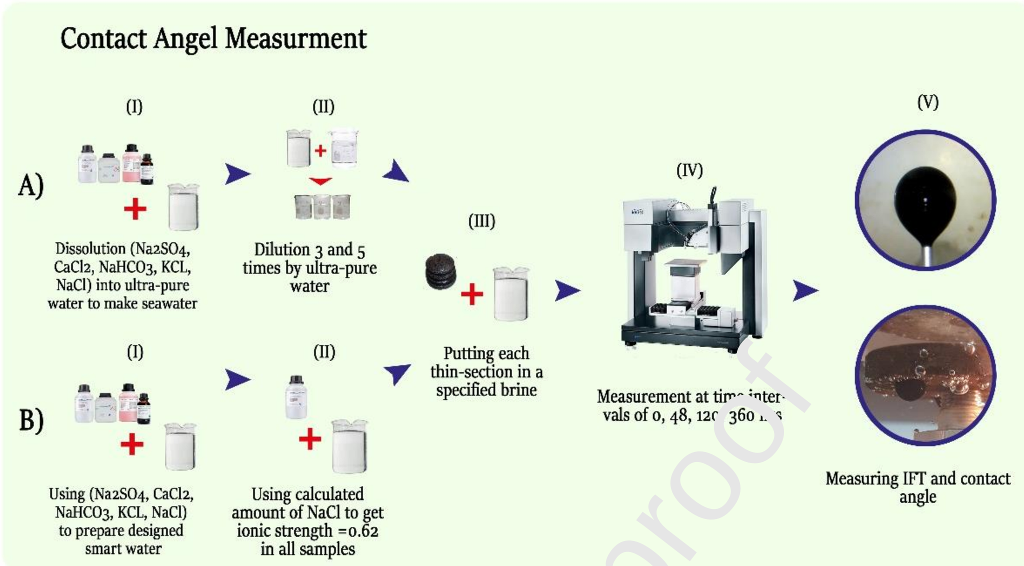

Figure 19: measurement process of IFT and WA determination

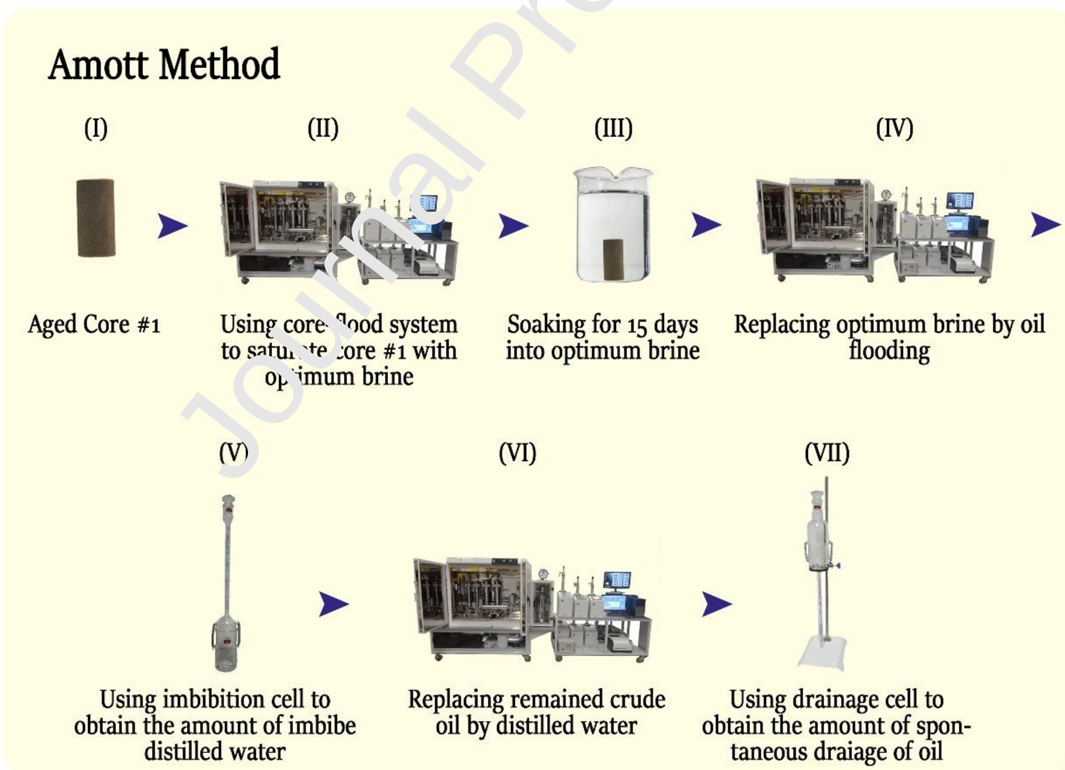

Figure 15: Amott method process to determine the pore scale WA in static condition

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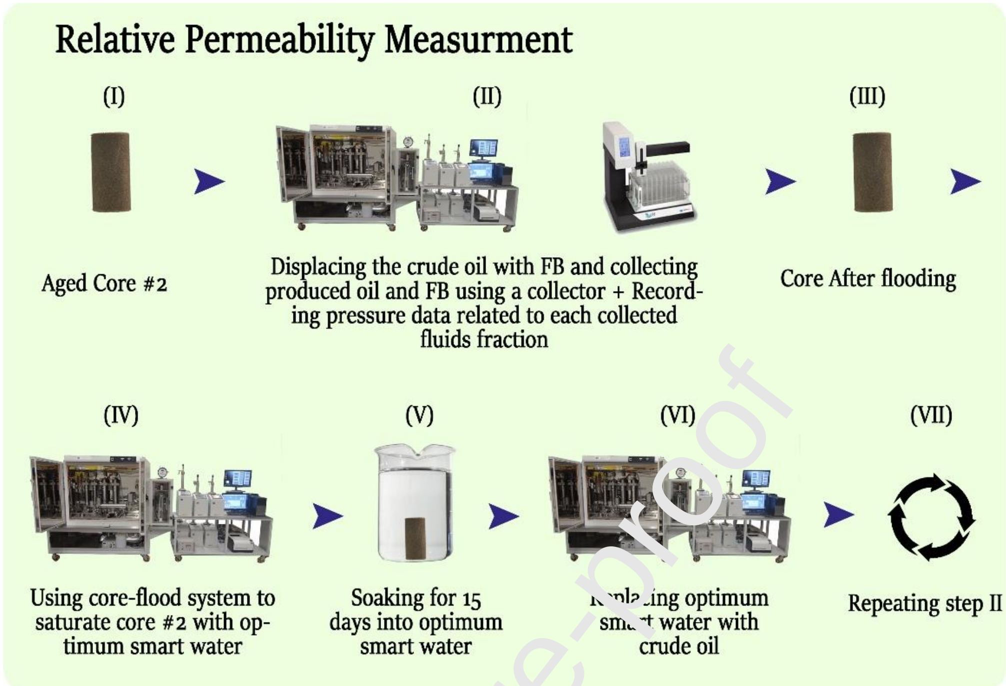

Figure 16: Relative Permeability measurement to determine pore scale WA in dynamic condition

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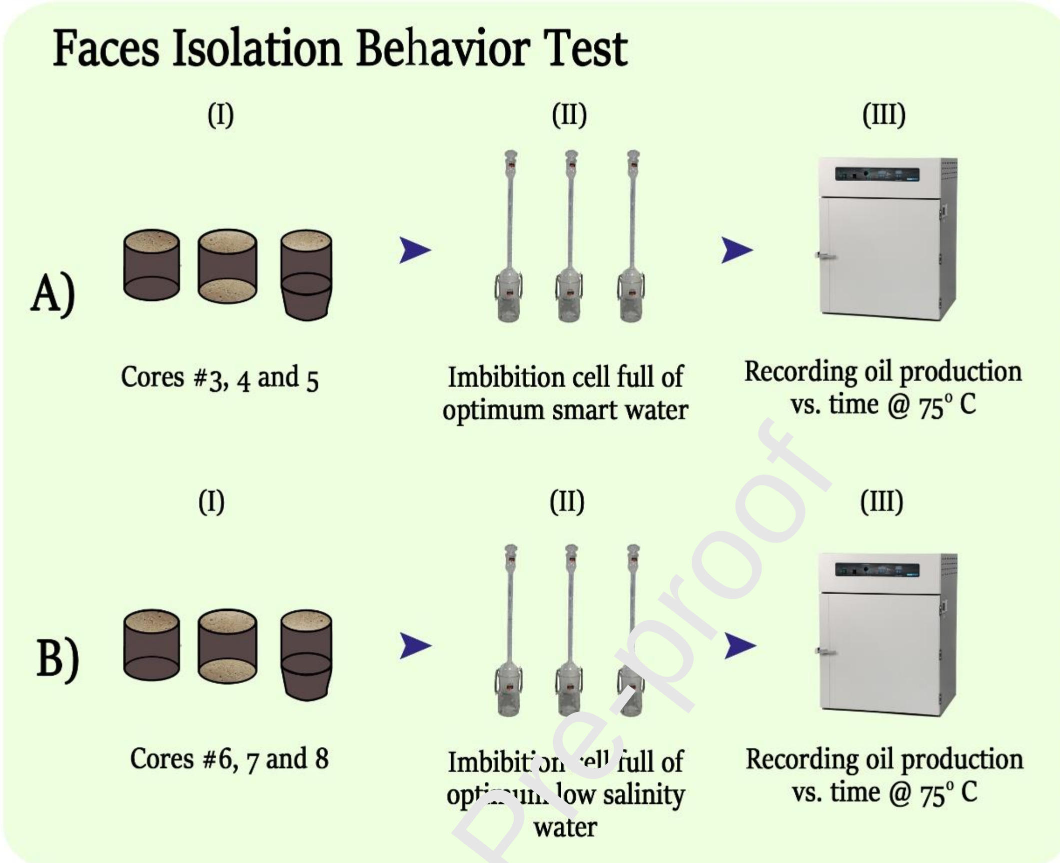

Figure 17: Procedure of core faces isolation behavior test

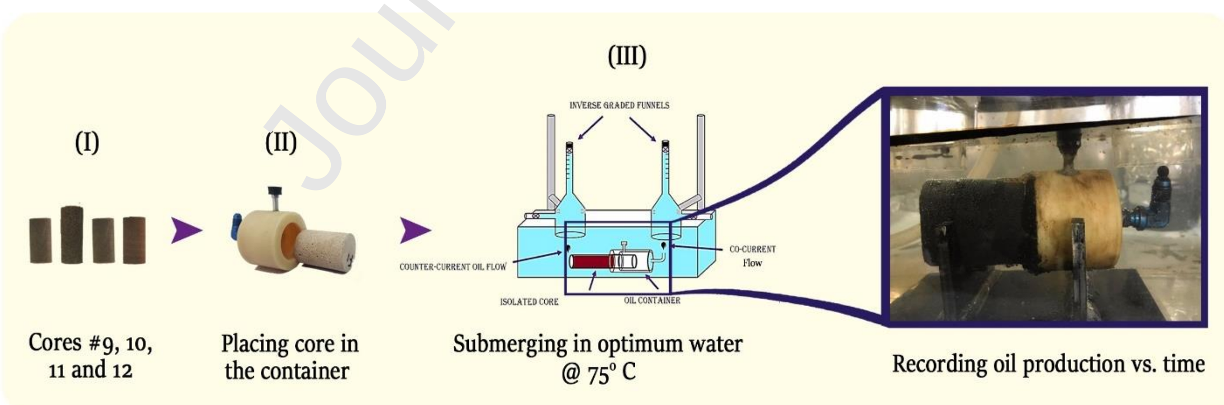

Figure 18: Procedure of core petrophysical properties effects on co- and counter-currents oil flow

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