



Smart Water

Master degree in oil and gas technology 10th Semester K10OG-4-F15

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Preface

This project is the core of the 10th semester for the students studying MSc Oil and Gas Technology in Aalborg University Esbjerg and is about Smart water or waterflooding in enhance oil recovery and also investigate this issue that Smart water will change wettability and surfactant will decrease IFT (may also change wettability) in this way both effects will play a role to recover additional oil.

This project has divided to three different parts that in first part attempts to investigate, analyze and evaluate Smart water method and different surfactants.

In second part that is data analyzing, has been used data from labratory and evaluate them in order to use them in process simulation.

The third part is about Process simulating and using UTCHEM software for simulation of smart water system.

In this project tried to explain all process and definitions in simple way and everybody with having general knowledge about engineering, could understand that.

All calculations and units are based on SI system.

Title of project: Smart water;

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1.1 Project introduction

The project is about introducing Smart water technology by investigating the method of enhance oil recovery. The ultimate objectivity in the project is to use Smart water method with a surfactant and changing wettability and IFT to recover maximum oil.

Smart water or water flooding is an interesting solution in oil and gas field, as companies are making research about it - whether it is a suitable way or not. The most important factor in this topic is the price of this technology, as companies want the cheapest solution when using surfactants and injections in reservoirs.

This project is a master thesis for a student studying MSc Oil and Gas Technology at Aalborg University in Esbjerg. The composition of the well describes its function, as well as the pressure, temperature, and mass flow. All written numbers and data is from the laboratory or from books. While working in the laboratory all calculations are in realistic conditions.

The learning objectives are as follows:

- To understand the Smart water process and Surfactants
- The ability to analyze and simulate the system



1.2 Problem definition

At the beginning, the main problem has to be considered and how it can be solved. What is Smart water and how can it be used in oil recovery, as well as, which types of surfactants are suitable and to analyze the final result.

In Enhance Oil Recovery (EOR) the main goal is to increase oil extraction from reservoirs, where there are several and different methods that can be functional, which are under investigation. EOR uses Smart water or water flooding to replace oil between rocks and stones by injecting one chemical liquid called surfactants in the oil field, which mixes together with water. The used surfactants in this method are different, as they have different parameters, and the selected surfactants have to be evaluated, whether it is suitable or not, and is it logical or not. To answering these questions, different kind of surfactants will be investigated based on the projects condition.

Different factors will be analyzed when the theoretical and practical investigation is done with the result from the laboratory and different experiments. This project will simulate and evaluate the UTCHEM software.

Other key aspects are:

- To investigate Smart water and surfactants
- Analysis of data (data is given from experiments)
- Modeling of process in UTCHEM program.

The aim in this project is to answer above questions and investigate Smart water technology in the best condition.



2. Enhanced Oil Recovery (EOR)

Enhanced oil recovery (curtailed EOR) is the use of a few strategies for expanding the measure of unrefined oil from an oil field.

As indicated by the US Department of Energy, there are three essential systems for EOR that are chemical injection, gas injection and thermal recovery.

Infrequently the term quaternary recovery is utilized to indicate to more theoretical, progressed, EOR systems. Utilizing EOR, 30 to 60 percent, or more, of the supply's unique oil can be extracted, in comparison with 20 percentage using primary recovery and 40-percentage secondary recovery. [Ref 1]



Figure 1 Enhanced Oil Recovery [Ref 2]

2.1 Techniques

As specified some time recently, there are three essential systems of EOR: chemical injection, gas injection and thermal recovery. Chemical injection, that can include the utilization of long-chained molecules titled polymers to rise the efficiency of waterfloods, can increment around one percent of EOR creation in the United States.



Gas injection, that uses gasses, for example, nitrogen, common gas or carbon dioxide (CO2), can build oil recovery producing almost 60 percent and Thermal recovery, that includes the presentation of warmth, expand oil recovery producing 40 percent.

In 2013, a strategy called Plasma-Pulse innovation was demonstrated into the United States from Russia. This procedure can rise oil recovery around 50 percent.

2.1.1 Chemical injection

This technique typically use for decline surface tension and release mobility and for applying, different chemicals and usually with dilute solutions.

With adding a dilute solution of a water-soluble polymer that can increase the viscosity of the injected water, we can increase the amount of oil recovery in some structure. Dilute solutions of bio surfactants such as rhamnolipids of surfactants such as petroleum sulfonates can be injected to lower the interfacial tension or capillary pressure that postponements oil droplets from moving through a reservoir.

Special constructions of water, oil, and surfactant, micro emulsions may be predominantly effective in this.

Generally, the critical factor in this application is cost. Each reservoir based on their formation and properties, have different absorption and this factor can play an important role in absorb or retain of surfactant. This problem cause to increase using of surfactants and it is not economy and suitable for companies. [Ref 1]



Figure 2 Chemical injection [Ref 3]



3. Waterflooding

This technology refer to use of water injection for increase of oil producing from reservoirs. This method also call secondary recovery and usually come after primary production and it done due to produce oil with use natural energy of reservoir (solution-gas drive, aquifer influx fluid, rock expansion, and gravity drainage). [Ref 4]

3.1 Rationale for waterflooding

As said before, the main reason of using water flooding is to increase of oil production. This is complete by "voidage substitution"—inject of water to expand the pressure to its starting level and keep up it close to that pressure. This water displaces oil from the pore spaces, but the effectiveness of this displacement is related to several parameters such as rock formation and structure and viscosity.

In oil fields (this project =North Sea), voidage replacement additionally has been utilized to direct extra surface diving. [Ref 4]

3.2 History

Salty water or brine was in the beginning of oil industry often produced from a well together with oil, the oil production decreased, as the rate of water production would grow. Usually, the water was set by putting it into nearby streams or rivers. The practice began in the 1920's by reinjections of the made water into porous and permeable subsurface formations, as well as the reservoir interval where the oil and water in the beginning came from. Reinjections of produced water had become a common oilfield practice by the 1930's.

The first reinjection of water happened in the Bradford oil field of Pennsylvania, Us, where the ''circle-flood'' was replaced with a ''line flood,'' and besides the ''line flood'' a ''five-spot'' well layout was used as well in the 1920's. In the production of wells, the two rows that were put on both sides of an equally spaced row of water-injection wells.

Water flooding technology and common practice is mostly established in America by 1940 and 1970. The onshore American oil industry was maturing by the 1940's, where the main production from its reservoirs had weakened severely, however, anywhere else in the world most reservoirs were in an early stage of main production. In America, drilling of closely spaced wells had been made too, to make the effects of water injections more significant.

The need of positioning the produced salty water alongside with oil, made water flooding a method that is reasonable and economical for growing recovery from oil fields. It was early noticed, that in most reservoirs the original oil in place (OOIP) was recovered in small percentage due to the main production period, which is caused by the depletion of the reservoirs' natural energy. The large quantity of oil that remained needed



to be produced by a recovery method. The early success of water injections helped improve the oilproduction period, as water flooding became the natural way to regain additional oil from reservoirs.

By water flooding's development, water becomes low-priced, as water is available from streams, rivers, oceans, and from wells drilled into aquifers. It is scientifically proved, that water has viscosity, density, and wetting properties because of the success by water flooding. As water flooding was the logical recovery process for most onshore oil fields in America, Russia, and China, some American offshore oil fields, as elsewhere in the world, were getting water injection. From the 1970's, many significant water-injection projects have been applied to oil reservoirs all over the world. [Ref 4]

3.3 Waterflooding considerations

Water displaces oil by unit displacement efficiency from a porous and permeable rock on a microscopic scale. At this level of analysis, it is most functional when water, oil, and flow measurements are made in laboratory on a small core-plug samples. To determine how good the water flooding will work, calculations of the effects of geology, gravity, and geometry must be included. The formula for complete water flood oil-recovery efficiency, where E_R is the product of three independent terms: [Ref 4]

$$E_R = E_D E_I E_A,$$

That

- E_D = the unit-displacement efficiency,
- E_I = the vertical-displacement efficiency,
- E_A = the areal-displacement efficiency.

However, the independence of these factors assumes not to be valid for real oil reservoirs.

The importance of technical and financial success of a water flood are oil properties, where the properties are viscosity and density. In a permeable medium, the fluids mobility is defined by its endpoint that is relative permeability divided by its viscosity. Therefore, a fluid with a low viscosity (\leq 1 cp) has a high mobility, though, if its relative permeability is very low. Likewise, a low-API crude oil (\leq 20°API) has a high viscosity and a very low mobility unless it is heated to high temperatures.

The temperature of waters viscosity is usually much lower or equal to the oil reservoir, where the ratio of water and oil viscosity is usually greater than 1:1. The ratio of water and oil mobility is important to the efficiency of the water/oil displacement process, as the recovery efficiency increases, the water/oil mobility ratio decreases.

More details about oil replacement by water injection are:



- Vertical displacement in a waterflood
- Macroscopic displacement efficiency of a linear waterflood
- Microscopic efficiency of waterflooding
- Areal displacement in a waterflood

3.4 Reservoir geology considerations

When evaluating a water flooding project, it is important to understand the reservoir rocks, and to reach this understanding it is by knowing the depositional environment at the pore and reservoir levels, perhaps also levels in between. The determination of the digenetic history of the rocks, then the determination of the structure, and the criticism of the reservoir to understand the interconnectivities among different parts of the reservoir – in particular the injector/producer connectivity. At the end, the characterization of water, oil, and rocks is needed to understand due to their control of wettability, residual oil saturation to water flooding, as well as the relative permeability of oil at higher saturations. Therefore, there should always be in the water flood evaluation team a developmental geologist.

The main geological evaluation in water flooding is to evaluate the nature and degree of heterogeneities that are in an exact oil field, as oil reservoirs are all heterogeneous rock formations, which can take many forms as in shale, anhydrite, or other impermeable layers. Interbedded hydrocarbon-bearing layers that have different rock qualities such as sandstones and carbonates, changing continuity, and environmental or digenetic changes that is caused by directional permeability trends. Fracture trends that developed due to regional tectonic stresses on the rock and the effects of burial and uplift on the particular rock layer. Fault trends that cause the connection of one part of an oil reservoir to adjacent areas, because they are either flow barriers or open conduits that allow unlimited flow along the fault plane. [Ref 4]



Figure 3 Reservoir geology [Ref 5]



A geological consideration is the structure of the reservoir, as well as how it affects the water flood process, because as the structure creates dipping beds at different angles, the interaction between the bed angle, gravity, and the oil/brine density. Furthermore, it affects the behavior of the relative vertical and horizontal flow. What can also be included is the structural thoughts whether the oil column has a primary aquifer or a covering gas cap – both can affect the probability of success in water flooding.

A technical team such as geologists and geophysicists must take into consideration such geological and structural aspects of a reservoir. To understand depositional environment and post-depositional digenesis, geologists use cores and routine-core-analysis data, where geologists are able to describe the reservoir's internal construction. Geophysicists use seismic data to distinguish between faults and trends in rock quality.

When using water flood, it is required to describe the quantitative evaluation, as well as different approaches may be used. The concept of the ''flow unit'' is how the volume of reservoir rocks affect the fluid flow, whether it is consistent or different from other rock volumes.

The evaluation process starts when a reservoir has been revealed, then a water flood will be initiated. The production and injection data will show the internal features of the rock volume that has been flooded, however, the data is critical, because the data relate to the interwell connectivity within reservoir, which either validates or causes change of the geoscientists' concepts.

Tracers can be injected during water flood to track injector/producer pairs. Other techniques include the use of specially drilled observation wells and 4D-seismic interpretations in order to track the way and shape of the higher-pressure water swept reservoir areas – they are centered on the injection wells.

3.5 Limitations of waterflood technology

By using water flooding, it can increase the volume of oil recovered from a reservoir, but it is not always the best solution, which can lead to confusing factors such as compatibility of the planned injected water with the reservoir's connate water, interaction between injected water and reservoir rock, removed oxygen, bacteria, and undesirable chemicals, and occurrence of radioactive materials. When producing a best oil reservoir, a petroleum engineer should take into consideration, that the water flooding should be analyzed in various ways such as technically and economically. [Ref 4]



3.6 Key points concerning waterflooding [Ref 4]

- Waterflooding is a common method in oil enhance recovery because water is low-cost and almost available in large volume and based on experiences because water is very effective at substantially, is very suitable for EOR and increase oil recovery.
- The efficiency of waterflooding depend on mobility ratio between water and oil and geology of the oil reservoir. Most of the time Waterflooding is effective and efficient because almost all reservoir rocks are either mixed-wet or water-wet. The depositional and diagenetic characteristics of a reservoir regulate major aspects of the oil/water transposition process. These characteristics can increase waterflood performance or have damaging effects on the WOR during the time. Frequently, the details of a reservoir's geology are not known till production wells start to produce injected water.
- Gravity effects that interplay between the density and gravity effects and the geologic layering of a reservoir. This parameter is important in waterflood because at reservoir circumstances, oil always is less dense than brine or injected water. This interplay can either hurt or help to waterflood performance comparative to a homogeneous system.
- Waterflooding is a process that characteristically takes several decades to complete. Therefore, continuous, routine field production and pressure data must be taken for analyzing and monitoring waterflood performance. There are opportunities to modify the original waterflood design and operating strategies based on analysis of the actual field production data. Sometimes, more expensive, special-data acquisition programs (3D- or 4D-seismic data) are run to support the evaluation process. A diversity of engineering tools have been established to analyze waterflood performance, extending from simple plots of field production data to full-field numerical-reservoir-simulation models.
- Waterflood can be modify and with the waterflooded intervals, changing the distribution of injection water among the injection wells and drilling additional wells at infill locations, and/or modifying the pattern style.
- Waterflooding can be used in offshore and onshore all over the world in different fields with different sizes.

4. Waterflooding in carbonate reservoirs [Ref 6]

Waterflooding contains the use of water injecting to relocate oil in a reservoir. Usually, oil recovery with refining volumetric sweep productivity via a variety of techniques and practices, containing in-fill drilling, improved reservoir description, high resolution reservoir simulation, advanced surveillance and monitoring and multidimensional wells.

Currently About 50% of world's known oil reservoirs are carbonate.





Figure 4 Waterflooding [Ref 7]

4.1 Considerations

In carbonate reservoir, whenever waterflooding is main considerationThe all factors and affected aspects should be evaluate.

Some Factors are constant and some of them are not changeable and also there are some properties that come from formation and structure of them. Those factors include:

- Permeability
- Wettability
- Porosity
- Contact angle
- Rock properties
- Fluid properties
- Fracture Intensity [Ref 6]

4.2 Water Modifications

As mentioned before, In waterflooding the water relocates oil from the pore spaces, but the efficiency of this displacement depends on several parameters such as rock characteristics and oil viscosity.



Since carbonate surfaces have positive charge, the water must be altered, generally by adjusting the salinity, which will affect the concentration of calcium ions (Ca2+), sulfate ions (SO42-), or magnesium ions (Mg2+).[Ref 6]

4.3 Low Salinity

Researches have found that with low salinity of injected water can change wettability of rock and increase oil recovery but it still is not clear why lower salinity water can do this.

For this issue there are three different theories:

• Rock dissolution:

Initially proposed by Hiorth et al., this theory clarifies the low-salinity effect by hypothesizing that the lower calcium concentration in low-salinity brine causes calcium carbonate from the rock to dissolve and start equilibrium with the brine. The adsorbed oil components are detached and the rock surface is purified more water-wet, whenever the calcium carbonate dissolves.

• Surface ion exchange:

At usual carbonate reservoir circumstances, rock surfaces have positive charge whereas the acidic components of oil have a negative charge, causing the rock to be mixed-wet or oil-wet.

If the determining anions in the water (SO42-) have a higher empathy to the rock surface than the acidic oil components, the anions are adsorbed and the oil is desorbed.

If the rock surface charge is reduced (because of low salinity), desorption of negative charged oily material is also simplified.

• In-situ surfactant formation:

The potential of in-situ surfactant generation requires a high pH and Hiorth et al. determined it to be a rare mechanism in waterflooding.

4.4 Advantages and Disadvantages

<u>Advantages:</u>

The main reasons that Waterflooding can be a suitable method for oil recovery are:

• Injected water is efficient to replace oil of light to medium gravity;



- water is easy to inject into oil field formations;
- water is cheap and almost available;
- In some aspects, if we compare waterflooding with other EOR methods it is obvious that maybe is an economic way and operation system is not so expensive.

Disadvantages:

Carbonate reservoir approach a badly-behaved in oil recovery, because about 80% of these geologic developments are oil-wet. In addition, Carboxylic acid anions have negative charged and it has to involve for positive charge in carbonate surfaces, hence producing oil-wet surfaces. As we know, because capillary pressure curves is mostly negative waterflooding cannot be successful in oil recovery. Consequently, surfactant/polymer waterflooding is suitable for some in the business to be a more practical alternative. [Ref 6]

5. Definition of theoretical terms

5.1 **Porosity**

Porosity is a measure of its capacity to hold a liquid. Numerically, porosity is the free space in a rock or stones divided by the total rock volume (Solid + space or openings).

Porosity is typically characterized as a rate of the total rock, which is taken up by pore space. For instance, a sandstone may have 8% porosity, this implies 92 percent is solid rock and 8 percent is open space containing water, oil or gas. and left 8 percent is about the minimum porosity that is obliged to make a proper oil well, however non-economic wells or poorer have less porosity.

In any case, sandstone is hard and seems extremely strong; it is similar to a wipe (a hard, incompressible wipe). Between the grains of sand, enough space exists to trap liquids like oil or normal gas! The gaps in sandstone are called porosity (from "permeable").[Ref 8]





Figure 5 sedimentary-rocks-sandstone-schematic [Ref 9]

The porosity is indicated as dark in the drawing. Gas, water or oil will fill these openings in the rock!

Subsequently, a poor-routed sandstones will have less porosity than a well-rounded one!

A well-rounded sandstone can be more interesting for geologist because these kind of rock can hold more amount of oil and gas!

Porosity (Φ) can be defined:

 $Porosity = \frac{Pore Volume}{Total Volume} = \Phi.$

Porosity define as a volume proportion so, is dimensionless and is generally reported as a part or percent. To evade disarray, especially when variable or changing porosities are included, it is regularly reported in porosity units (1 PU = 1%).

A few volume definitions are obliged to represent porosity:

Total volume of rock	$= V_T \text{ or } V_{rx}$
Volume of mineral phase	$= V_g \text{ or } V_m$
Volume of pores or openings	$= V_{\rm por}$



From these we can characterize the different sorts of porosity experienced:

Total porosity	${\Phi}$	$= V_{\rm por} / V_T$
Effective porosity	Φ_{p-e}	$= V_{p-con}/V_T$
Ineffective porosity	$\Phi_{p-\mathrm{iso}}$	$= V_{p-iso}/V_T$
Crack or fracture porosity	Φ_{fx}	$= V_{fx} / V_T$

5.2 Permeability [Ref 10]

The permeability factor in rock is a term that define as resistance to the stream of a liquid through a rock. The rock called low-perm whenever needs lots of pressure for press the fluid through the rock and if liquid goes through the rock effortlessly; its call high-perm or the rock has high permeability.



Figure <u>6</u> Permeability measures how easily fluid passes through a rock [Ref 11]

In oil production, Permeability expressed by the unit that is millidarcys. One millidarcy is 1/1000 of a Darcy. One Darcy is a high amount of permeability!



Most of reservoirs that can be used for oil and gas production have permeability around ten to several hundred millidarcys.

The increasing amount of US gas production in the last 10 years is coming from shale gas wells that has a big amount of porosity (much more than sandstone), but it has enormously low permeability. Therefore, it means shale is poor producer of hydrocarbons. For over a hundred years, gas has been produced from shale but quantities were small. Allowing for improved shale gas expansion, two things have changed the situation. Based on these concepts, oil and gas companies have been allowed to persuade more permeability into shale gas rock artificially:

- *Horizontal Drilling* The broad adoptance of Horizontal Drilling method, in which the drill bit is made to turn from the vertical to the horizontal (a 90-degree turn), where it can keep to drill horizontally through the structure. The horizontal track can be as much as a mile! [Ref 20]
- Advances in Hydraulic Fracturing (fracking) Fracking is not another innovation; it has been around well more than 50 years (regardless of what you may hear on the news). On the other hand, propels in fracking systems in horizontally gaps, especially in shale structure, has prompted a massive increment in shale gas producing. These new methods permit the oil and gas administrator to render low-permeability shale reservoirs more permeable, by artificially bringing little cracks into the structure.

Permeability		Pervi	ous		Semi-Pervious		Impervious						
Unconsolidated Sand & Gravel	Well Gr	Sorted avel	Well or Sa	Sorte and &	d Sand Gravel	Sand Very Fine Sand, Silt, ravel Loess, Loam							
Unconsolidated Clay & Organic					Pe	at	La	yered	Clay	ι	Inweath	nered C	Clay
Consolidated Rocks Highly Fractured Rocks		Oil F F	Reserve Rocks	oir.	Fr Sand	esh Istone	Fr Lime Dole	esh stone, omite	Fresh	Granite			
к (cm ²)	0.001	0.0001	10 ⁻⁵	10 ⁻⁶	10 ⁻⁷	10 ⁻⁸	10 ⁻⁹	10-10	10 ⁻¹¹	10 ⁻¹²	10-13	10-14	10 ⁻¹⁵
κ (millidarcy)	10+8	10 ⁺⁷	10+6	10+5	10,000	1,000	100	10	1	0.1	0.01	0.001	0.0001

Source: modified from Bear, 1972

Figure 7 Permeability measures how easily fluid passes through a rock [Ref 12]



Permeability is part of the proportionality constant in Darcy's law. This factor relate to flow rate and physical properties of liquid (viscosity), to a pressure gradient applied to the porous media:

$$v = \frac{\kappa}{\mu} \frac{\Delta P}{\Delta x}$$

Therefore:

$$\kappa = v \frac{\mu \Delta x}{\Delta P}$$

where:

v is the superficial fluid flow velocity through the medium (i.e., the average velocity calculated as if the fluid were the only phase present in the porous medium) (m/s)

 κ is the permeability of a medium (m²)

 μ is the dynamic viscosity of the fluid (Pa s)

 ΔP is the applied pressure difference (Pa)

 Δx is the thickness of the bed of the porous medium (m)

In normally happening materials, permeability values range over many orders of scale.

5.3 wettability

Wettability can define as tendency of one liquid to spread on, or hold fast to, a solid surface in the vicinity of other immiscible liquids. Wettability refers to the interaction between liquid and solid phases. In a reservoir, solid phase can be the rock mineral accumulation and the fluid phase can be oil, water or gas.

Wettability is all around branded by the contact angle of the liquid with the solid phase. Interfacial tension (σ) is the energy per unit area (force / distance) at the surface between phases. It is usually expressed in milli Newton / meter or dynes / cm.

Adhesion tension (γ) is expressed as the difference interfacial tensions between two solid and liquid phases. When γ is negative, that means denser phase (water) better wets the solid surface (and vice versa). An adhesion tension of 0.0 shows that both phases have equal empathy for the solid surface.

As we know at least 3 groups of forces acting on the fluids whenever 2 or more fluids are present, and it can effect on hydrocarbon recovery.





Figure 8 Wettability [Ref 14]

- γ = adhesion tension
- θ = contact angle between the oil/water/solid interface measured through the water
- σ_{os} = interfacial energy between the oil and solid
- σ_{ws} = interfacial energy between the water and solid

 σ_{ow} = interfacial energy (interfacial tension) between the oil and water

Attractive forces between fluid and rock draw the wetting phase into slight pores. Wetting phase fluid frequently has low mobility.

Many hydrocarbon reservoirs are either totally or partly water-wet. Attractive forces describe the lower limit to irreducible wetting phase saturation.



Figure 9 Wettability [Ref 14]

if water better wets the rock surfaces reservoir rock is water wet and The rock is water wet in these conditions:



$$\label{eq:states} \begin{split} \sigma_{ws} &\geq \sigma_{os} \\ \gamma < 0 \mbox{ (adhesion tension is negative)} \\ 0^\circ &\leq \theta \leq 90^\circ \end{split}$$

When θ is close to 0° , the rock is reflected to be "strongly water wet".

Between fluid and rocks repulsive forces effect nonwetting phase to inhabit largest pores. Nonwetting phase does not better wet the solid rock surface.

In hydrocarbon reservoirs, natural gas is never the wetting phase.

The most mobile fluid is frequently nonwetting phase, especially at large nonwetting phase saturations.



Figure 10 Wettability [Ref 14]

If oil better wets the rock surfaces, reservoir rock is oil-wet. The rock is oil wet in these conditions:

$$\label{eq:sigma_os} \begin{split} \sigma_{os} &\geq \sigma_{ws} \\ \gamma &> 0 \mbox{ (adhesion tension is positive)} \\ 90^\circ &\leq \ \theta \leq \ 180^\circ \end{split}$$

When θ is close to 180°, the rock is well-thought-out to be "strongly oil wet"



Figure 11 Comparison of water wet and oil wet rocks [Ref 14]



Wettability is categorized by these variations:

- Strongly water or oil wetting.
- Neutral wettability no better wettability to either oil or water in the pores.
- Fractional wettability reservoir that has local areas that are strongly oil-wet, while most of the reservoir is strongly water-wet happens where reservoir rock has changeable mineral composition and surface chemistry.
- Mixed wettability smaller pores filled with water and are water wet, while larger pores filled with oil and are oil wet. Residual oil saturation is low happens where oil with polar organic compounds occupies a water-wet rock saturated with brine.
- Wettability differs with surface roughness, so the wettability of a rock will differ with size, grain shape, and rounding.



Figure 12 Roughness or angularity of grains affects wettability [Ref 14]

The present measuring technique uses a modern contact angle goniometer and is called the static sessile drop method.

The contact angle goniometer is an optical subsystem which catch the profile of an immaculate fluid on a solid substrate. The angle shaped between the solid and liquid interface and the fluid/vapor interface is the contact angle. More experienced frameworks utilized a magnifying instrument optical framework with a backdrop illumination. Current-era frameworks utilize high determination cameras and programming to catch and investigate the contact angle.

The dynamic sessile drop system is like the static sessile drop but obliges the drop to be altered. A typical sort of element sessile drop study decides the biggest contact angle conceivable without expanding its liquid and solid interfacial territory by including volume dynamically. This maximum angle is the propelling angle. Volume is removed to deliver the littlest conceivable angle, the retreating angle. The distinction between the progressing and retreating angle is the contact angle hysteresis.



5.4 Surfactants [Ref 15]

A surfactant is fleetingly defined as a material that can greatly reduce the surface tension of water when used in very low concentrations.

Surfactants are usually organic compounds that are amphiphilic, and it means they have both hydrophobic groups (their tails) and hydrophilic groups (their heads). Thus, a surfactant has both parts that are water insoluble (or oil soluble) component and a water-soluble component.

In the mixture of oil and water, surfactants will diffuse in water and adsorb at interfaces between air and water or at the interface between oil and water.

The water-insoluble hydrophobic group can spread out of the bulk water phase, into the oil or into the air phase, whereas the water-soluble head group remains in the water phase.

Reliant on on the nature of the hydrophilic part the surfactants are categorized as Cationic, Anionic, Amphoteric or Nonionic.





Anionic surfactants

When the hydrophilic part of the surfactant contains of a negatively charged group like a sulphate, sulphonate, or carboxylate the surfactant is named anionic. Basic soaps can be a good example of anionic surfactants. In the last 50 years, many soaps have been replaced with more efficient substances such as Alkyl sulphonates, alkyl sulphates, and alkyl benzene sulphonates. Anionic surfactants are sensitive for water hardness.



Nonionic surfactants

A surfactant with a non-charged hydrophilic part, for example ethoxylate, is non-ionic. These materials are suitable for cleaning purposes and are not sensitive for water hardness. They have a extensive use in cleaning detergents and include groups like polyglycosides, fatty alcohol and alcohol ethoxylates.

Cationic surfactants

For this group the hydrophilic part is positively charged for example with a quaternary ammonium ion. This category of surfactants has no wash activity effect, but secures to the surfaces where they might provide antistatic, softening, anti-bacterial, soil repellent or corrosion inhibitory effects. The most typical applications are for antistatic and softeners. The cationic surfactant also known by DADMAC.

Amphoteric surfactants

For the amphoteric surfactants the charge of the hydrophilic part is organized by the pH of the solution. It's means that they can performance as anionic surfactant in an alkalic solution or as cationic surfactant in an acidic solution.regular increase in TMP after each backwash, however, the backwash process cannot always remove accumulation of foulants alone. Chemical cleaning are needed.

5.4.1 Effect of surfactants on wetting

Control of liquid dispersal over solid surfaces require many technological processes. When a drop is located on a surface, it can entirely wet, not wet the surface or partially wet.

By decreasing the surface tension with surfactants, a nonwetting material can become completely or partially wetting. The extra free energy (σ) of a drop on a solid surface is: [Ref 19]



$$\sigma = \gamma S + PV + \pi R^2 (\gamma_{\rm SL} - \gamma_{\rm SV})$$

That:

- γ = liquid-vapor interfacial tension
- γ_{SL} = solid–liquid interfacial tension
- γ_{SV} = solid-vapor interfacial tension
- S = area of liquid-vapor interface
- P = excess pressure inside liquid
- R = radius of droplet base

Based on this equation, the excess free energy is diminished when γ declines, γ_{SL} declines, or γ_{SV} rises. Surfactants are suck up onto the liquid–vapor solid–liquid, and solid–vapor interfaces, which adjust the wetting behavior of hydrophobic substances to decrease the free energy. When surfactants are suck up onto a hydrophobic surface, the polar head groups expression into the solution with the tail pointing outward.

In more hydrophobic surfaces, surfactants might form a bilayer on the solid, triggering it to become more hydrophilic. The dynamic drop radius can be measured as the drop begins to spread. Therefore, the contact angle changes based on the following equation:

$$\cos \theta(t) = \cos \theta_0 + (\cos \theta_\infty - \cos \theta_0)(1 - e^{\frac{-t}{\tau}})$$

- θ₀ = initial contact angle
- θ_∞ = final contact angle
- τ = surfactant transfer time scale

As the surfactants are sucked up, the solid-vapor surface tension increases and the edges of the drop become hydrophilic. Therefore, the drop spreads.

6. Experiments

In this part tried to achieve real results from different experiments in laboratory and use them for analyzing and at the end for project simulation.



For all experiments, synthetic seawater has been used. For making seawater, this followings considered:

• composition of seawater:

(Salinity = 35)						
Component	Concentration (mol/kg)					
H ₂ O	53.6					
cī	0.546					
Na [⁺]	0.469					
Mg ²⁺	0.0528					
so ₄ ²⁻	0.0282					
Ca ²⁺	0.0103					
ĸ⁺	0.0102					
CT	0.00206					
Br	0.000844					
BT	0.000416					
sr ²⁺	0.000091					
F	0.000068					

Total Molar Composition of Seawater (Salinity = 35)

Figure 14 Seawater compossition [Ref 17]

• Concentration or Salinity of seawater:

On average, seawater in the world's oceans has a salinity of about 3.5% (35 g/L). So base on this concentration, the final mixture is mixture of 500 ml water and 17.5 g sea salt.





6.1 Aqueous stability

The main aim of this experiment is to find a stable surfactant for waterflooding. Due to find a stable surfactant, different kinds of surfactants have been test and finally two surfactants are chose. For finding suitable surfactant, the mixture of seawater and surfactant has to stand on 60 C^{0} .

In this project, one anionic (SDS) and one cationic (P 50-70) [Appendix 1] surfactants have been chose and for stability test, the mixture of each surfactants and seawater with concentration of 2% stood 2 days on oven with temperature of 60 C⁰.



Figure 14 mixture of Seawater and Surfactant in oven on 60 C⁰

After 2 days, both surfactant mixtures were stable and homogeneous and could use in other experiments.

6.2 Contact angle for wettability

For this part, different mixtures with different concentrations tested and with camera, contact angle (θ) calculated.

At the beginning, for making chalk surface one simple technique used that with one adhesive tape and $CaCo_3$ powder one thin layer have been made. This method was not functional because surfactant solved the layer and camera was not able to recognize the drop. So One thin layer of chalk ($CaCo_3$) selected as a main surface and for each mixture one drop evaluated separately.





Figure 15 Contact angle measurement

These data will be use in calculation of surface tension.

6.3 Surface Tension

The main goal of this experiment is measuring of Surface Tension due to select a specific concentration of surfactant. This work has been done by Du Nouy ring method with different versions of synthetic seawater (SSW).

The main equation is:

$$\gamma = \frac{m.\,g.\,cos\theta}{4\pi.\,R}$$

That:

 γ = Surface tension (N/m) m = mass (g) θ = Contact angle R = Du Nouy-rings radius (m)

For this part, Surface tension in different mixtures with different concentration has calculated.



Seawater

 $\cos \theta$ for water is 1,

$$\gamma = \frac{0.65*10^{-3}kg*9.81*1}{4\pi*9.75*10^{-3}m} = 0.05207 \text{ N/m}$$

For SDS (Sodium dodecyl sulfate) in different concentration

In mixture of 2% seawater and SDS, $\theta = 45^{\circ}$ and if this mixture will be more diluted θ also reduced. In this part for SDS:

 $\theta = 37^{0}$ in 2D (two times diluted), $\theta = 29^{0} 10$ D (ten times diluted) and $\theta = 20^{0} 20$ D (twenty times diluted)



For P 70-50 in different concentration

In mixture of 2% seawater and P 70-50, $\theta = 72.5^{\circ}$ and if this mixture will be more diluted θ also reduced. In this part for P 70-50:

 $\theta = 65^{0}$ in 2D (two times diluted), $\theta = 53^{0}$ 10D (ten times diluted) and $\theta = 42^{0}$ 20D (twenty times diluted)





6.4 Interfacial tension (IFT)

Interfacial tension is the force that grips the surface of a particular phase together and is normally measured in dynes/cm and this force exists when two phases are present. These phases can be gas/oil, oil/water, or gas/water.

In this part, IFT in different mixture of oil and seawater and 2 different surfactants (SDS and P70-50) has been calculated.



Figure 16 Different mixtures for IFT measurment



The main difference between IFT and surface tension is the places where it occurs. Surface tension is defined to a single liquid surface, whereas the interfacial tension is defined to the interface of two immiscible liquids.

After measurement of IFT in three different scenario base on these two graphs we can say that adding surfactant increase IFT.





Based on IFT calculations, in P70-50 maximum IFT happened in point 20 dynes oer centimeter and this number can be use in our simulation.

As conclusion, based on results and graphs we can say that adding surfactants increased IFT and Wettability and reduced surface tension.

7 Process simulation with UTCHEM software

1

UTCHEM is a three-dimensional chemical flooding simulator and in this part, tried to simulate the data that have gotten from experiments [Appendix 4] and evaluate adding P50-70 surfactant with 2% concentration.

⊢ ∰® –		
	Reservoir Description	Biodegradation
	Output Options	Recurrent Data
	Reservoir Properties	Run
	Physical Properties	Output
	Geochemical Data	Quit
2 . 0 . 0		

Figure 17 UTCHEM's main window



7.1 Reservoir description

Run number and Title		Number of Components	
Γ	Run number MAIN.P	Total number of components	6
	Title and run description	Number of organic components	0
	MASTER THESIS, ALIREZA POURIMANI	Number of water/oil tracers	0
SURFACTANT FLOODING		Number of oil/gas tracers	0
Run Options		Number of geochemistry components	0
	Model Options	Number of nel components	n
	Grid Description	Name of compon	ents
	Gridblock Sizes	Units of water/oil to	acers
		Components Pres	sent

In the first step in this program, reservoir description should be consider.

Figure 18 Reservoir Description

The title of project and number of components have to write here. As you can see for components part, 6 components considered that 3 of them (Oil, Water, Surfactant) are our main components, two components are from reservoir (Chloride and Calcium) and other component is an option for other surfactant or polymer.

Other important adjustment in this part is Model Option.

Model Options				
First simulation run Restart simulation Time Step Constant Time Step Automatic Time-Step by relative change of first 3 components erelative change of all components change in concentration of all components	Reaction Options			
Numerical Methods Single point upstream weighting Two point upstream weighting Total variation diminishing third order	Coordinate System Cartesian Cartesian (Variable-width gridblock) @ Curvilinear			
Using concentration well model Using capacitance model Using dual porosity model	 ☐ Including tracer reactions ☐ Using 2nd order time approximation ☐ Gas is present ☐ Foam option is used ☐ Including temperature effect 			

Figure 19 Model Option



As you can see in Figure 19:

<u>FIRST SIMULATION RUN</u> have been chose and for <u>TIME STEP</u> part, all components selected. For <u>NUMERICAL METHOD</u>, total variation diminishing third order has been chose.

For <u>REACTION OPTION</u>, the first option has been chose because in this project, using gel did not consider and or <u>COORDINATE SYSTEM</u> curvilinear condition has been chose.

7.2 Output Option

For this part, the final data the want to export from UTCHEM has to be consider.

Print Time Unit		Max Injection Time Unit		
⊖ day	5	pore volumes	🔿 days	pore volumes
Output Files Components :	Component Print * Water Oil Y Surf. Y heck print with " Y " pressure saturations onent concentration us phase concentration itance/fraction proper operties	n for biodegradation	□ Geochemistry properties □ Temperature □ Aqueous phase tracer concentration □ Component concentration in each □ Phase viscosity □ Phase viscosity □ Relative Permeability □ Capillary number/residual saturation □ Effective salinity □ Properties related to hyteresis □ Properties related to foam	on at observation points phase on/interfacial tension wass transfer
Profile Print Options	rption Z direction phase flux	es	Permeability reduction factor, poly equivalent shear rate Phase environment indexing	mer viscosity, and Tracer Concentration

Figure 20 Output Option

As you can see in Figure 20, after simulation we can extract Phase pressure, Phase saturations and component concentrations and for Maximum injection time, we can export temperature, phase viscosity and relative permeability.



7.3 Reservoir properties

Miscellaneous Input		Initial Gas Sa	turation / Organic Concentrations			
Ma	ximum simulation time	1.15		Initial	Gas Saturation	
Ro	ck compressibility	.0000043		Initial	Oil Composition	
Re	ference pressure	5000	Property Mod	ification		
Porosity and I	Permeability			Flag For P	roperty Modification	
					Porosity	
	Flag For Porosity and Permeabilit	y		X Direc	tion Permeability	
	Porosity			Y Directiv		
	X direction permeability			Z Direc	tion Permeability	
	Y Direction Permeability			Initial V	∀ater Saturation	
	Z Direction Permeability			Initial	Gas Saturation	
Initial Reserve	pir Properties		Brine Propert	es .		
	Flag For Initial Reservoir Propertie	es				
	Depth of Top Layer		Ini	tial brine salinity	.62	7
	Initial Pressure		Bri	ne divalent cation		
	Initial Water Saturation		co	ncentration	.13	3
	Initial Aqueous Phase Composition	ns				
						1
					OK	Cancel

In this part, the reservoir details have to be consider.

Figure 21 Reservoir properties

As you can see in Figure 21, maximum simulation time set on 1.15 days or 30 hours. For Rock compressibility and pressure, Figure 22 can be use that 4.3 and 5000 has been chose.

Overburden Pressure (psia)	PV (cm ³)	Bulk Volume (cm ³)	Porosity (%)	Pressure (psia)	Rock Expansivity	Rock Compressibility (10 ⁻⁶ PV/PV/psi)
200	3.420	20.530	16.66	9,800	0.0000	15.1
1,000	3.379	20.498	16.48	9,000	0.0120	12.5
2,000	3.337	20.447	16.32	8,000	0.0243	10.2
3,000	3.303	20.413	16.18	7,000	0.0342	8.2
4,000	3.276	20.382	16.07	6,000	0.0421	5.8
5,000	3.257	20.367	15.99	5,000	0.0477	4.3
6,000	3.243	20.353	15.93	4,000	0.0518	4.0
7,000	3.230	20.340	15.88	3,000	0.0556	5.3
8,000	3.213	20.323	15.81	2,000	0.0605	11.3
9,000	3.177	20.329	15.63	1,000	0.0711	20.9
9,500	3.144	20.254	15.52	500	0.0807	

Figure 21 North sea's reservoir conditions [Ref 18]



For porosity and permeability, variable condition has been chose. For initial reservoir properties, the data that come from North sea reservoir properties has been used. [Appendix 2]

Flag For Ir	nitial Reservoir Properties	
Depth	of Top Layer	
	O Single value	
	 Depth of top gridblock and the reservoir dip angles 	
	Depth of each gridblock	
Initial I	Pressure	
	O Single value for all gridblocks	
	O Specified Depth	
	Each gridblock is specified	
Initial V	₩ater Saturation	
	O Single value for all gridblocks	
	Constant value for each layer	
	Each gridblock is specified	
Initial /	Aqueous Phase Compositions	
	Only anion and cation in aqueous phase	
	Constant value for whole reservoir	
	O Constant value for each layer	
	O Initial value for each gridblock	

Figure 22 Initial reservoir properties

And for brine properties 2 parameters have to consider that are salinity and concentration

.630

Figure 23 Brine properties

The unit of those number is meq/ml of water.



7.4 Phisical properties

The most important part In this section is IFT or interfacial tension that calculated in experiments and other parameters set by data sheet that come from Surfactant description and components description.

Concentration (volume fraction)	Organic Mass Transfer
Dil at plait point	Flag for Organic Dissolution
Type II (+) 0 Type II (-) 1	Organic Dissolution Data
Critical micelle (CMC) .0001	Residual saturation/Relative permeability
Using original Hand's rule	O Not depend on capillary number
O Using modified Hand's rule	Depend on capillary number Depend on trapping number
Phase Behavior Type	Capillary desaturation curve parameter
Using binodal curve	aqueous 1865
O Using solubilization ratio	oleic 28665.46
Phase Behavior Data	microemulsion phase 364.2
Interfacial Tension	Residual saturation/Relative Permeability
Healy and Reed's correlation	
○ Huh's correlation	Model and Type Selection
Interfacial tension parameters	AT low capillary number
Log 10 G (dynes/cm)	AT high capillary number

Figure 24

Phisical properties

7.5 Recurrent data

In this part, data about Well and boundary condition has to be consider.

Boundary Condition	Well Information	
☐ Boundary is specified	Well ID, Location,	
Saturated zone is modeled	Rate, conc. (Injectors)	
U vadose zone is modeled		
○ Saturated and Vadose zone are modeled	Rate, Pwi (Producers)	
Boundary Condition	Time Interval and Step	
	Cumulative injection time	.124
Number of total wells (including 2	Output frequency (Unit : Pore volumes)	
Radius model	Profile (PROFIL)	.124
C Padu and Odeb model	Prod. (PROFIL)	.124
	Prod. histories (HIST01,)	.01
· Peaceman model	Individual profile (SATP,)	.124
Min and Max Time Step Units	Restart (RESTAR)	1
O Days © Courant numbers	Time Step Option	
	Well Data Changes Information	
Number of actual wells	Well Data Change	





7.6 Run

MAIN.P				
TIME, DAYS 1.389 6.370 20.96 61.74 122.8 187.2 250.2 284.3	STEP 50 100 200 250 300 350 400	TIME STEP 0.5797E-01 0.1573 0.4854 1.211 1.293 1.273 1.080 0.9823	, DAYS COURANT 0.4898E-01 0.4892E-01 0.482E-01 0.4951E-01 0.5002E-01 0.5005E-01 0.4248E-01 0.3757E-01	NO.

After all calculation and data analyzing, with press of <u>RUN</u> button, all tables and plots are accessible.

Figure 26 Runing



Figure 27 Output files



As you can see in Figur 27 after running of data some file will export and these files are our final data about our simulation.

In overal section, all well's data and final calculation about running ti,e exist. Also in this section we can extract our data as an exle file or plot them [Appendix 3].

The Plots are :





This graph shows the variation of Pressure in different volumes of surfactant and it pressents adding surfactand cause to rise of pressure and after the point (0,6), the rate of pressure will be steady and It means the reservoir is in saturation condition and adding more surfactant can not increase the pressure.



This graph also is about variation of pressure but during the times and different days and similar to previous graph presents how pressure change in different days.





These two graphs show variation of cumulative oil per volumes (PV) and per days. Based on this calculation, adding surfactant increas amount of oil production.

The numbers of vertical axes present percentage of OOIP (Orginal oil in place).







These two graphs also show variation of cumulative oil per volumes (PV) and per days but in different unit that is BBLs (barrels).







These two graphs show overal rate of injection per volumes (PV) and per days and it is clear during the time, this rate will be increase.







These two graphs show overal rate of production (barrel per day) in different volumes (PV) and also different days.

Based on these two graphs, overal production rate is constant but after some days this rate will increase.







These two graphs presents amount of fluid injection during the time also different volumes.

If there are big variation between some points, it is natural because in some points amount of surfactant and injected water are different. In some points because of process pressure this amount should be lass than normal time.







These two graphs shows rate total fluid production after water injection in different volume and different days. Again same previous graphs, there different points that is because of reservoir properties and process conditions.







These two graphs shows rate of surfactant cumulative injection per volume and per day. The unit is FT3 or cubic feet and based on graphs, this parameter at the beginning is 0 and during the time with increase of surfactant injection will increase.





These two graphs shows rate of surfactant cumulative in production. As graphs say, at the begginig and after some days there is no surfactant but after one point this this rate suddenly increase and in durring the time this increase will continue.







These two graphs present amount of surfactant retained after production. This amount also during the time increase and after high rise in peak of production, will change to stady and constant rate.



8 Conclusion

In this project tried to explain main concept of smart water and evaluate different methods in this technology.

After data analyzing that should be based on theory part, some important experiments had been done. These experiments can explain different parameters in order to evaluate water flooding.

For experiments, two different surfactants chose and all calculation done for both of them. The main reason of this selection was comparison between Anionic and Cationic surfactants.

In experiments part, two essential parameters (wettability and Interfacial tension) considered and showed us with adding surfactant these two parameters also changed.

These parameters analyzed in different concentrations and based on surfactant's behavior, all calculations have been done logically.

For simulation part, cationic surfactant was under consideration and according to final simulation, we can estimate rate of injected surfactant, production rate, surfactant retained and other calculations.

Based on simulation graphs, adding surfactant at the beginning cannot increase oil production and after some days with increase pressure in reservoir and becoming saturated, oil recovery will increase.

With data about amount of recovered oil and surfactant this issue should be consider that using this surfactant is functional or not that in this project with using 1% surfactant was acceptable and just related to geology properties. With exact data and details about reservoir properties, this estimate would be more real and functional.

Another important issue is environmental impacts. Using chemical materials and inject to the reservoirs can change many parameters in seabed and effect on animals life. Retained surfactant also has to consider because based on materials and properties can be harmful for ecosystem.



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APPENDIX – **Data sheets**

[Appendix 1] surfactant's properties used from produced company

http://www.accudynetest.com/polytable_03.html?sortby=contact_angle

Molar mass	variable
Density	1.38 g/cm ³ (20 °C), ^[2] amorphous: 1.370 g/cm ^{3,[1]} single crystal: 1.455 g/cm ^{3[1]}
Melting point	> 250 °C, ^[2] 260 °C ^[1]
Boiling point	> 350 °C (decomposes)
Solubility in water	practically insoluble ^[2]
Thermal conductivity	$0.15^{[3]}$ to 0.24 W m ⁻¹ K ^{-1[1]}
Refractive index (n _D)	1.57–1.58, ^[3] 1.5750 ^[1]
The	ermochemistry
Specific heat capacity (C)	1.0 kJ/(kg·K) ^[1]



Source ^[1]	
Water absorption (ASTM)	0.16
linear expansion coefficient (α)	7×10 ⁻⁵ /K
Vicat B	82 °C
Glass transition temperature (Tg)	67 to 81 °C
notch test	3.6 kJ/m ²
Elastic limit	50-150%
Tensile strength(σ_t)	55–75 MPa
Young's modulus (E)	2800–3100 MPa

[Appendix 2]

 $https://www.gov.uk/government/uploads/system/uploads/attachment_data/file/197333/TR_SEA2_Geology.pdf$







[Appendix 3]

North



0	0	0	0	0	0	0	0	0	0	0	0	L.71E-05	2.62E-05	1.05E-05	5.42E-05	6.6E-05	7.8E-05).44E-05	.000111	.000127	.000141	.000154	.000166	.000178	.000193	.000209	.000224	.000241	.000256	.000268	.000279	.000292	.000304	.000317	0.00033	.000343	.000355	.000366	.000376	.000386	.000397	.000411	.000426	.000442	0.00046	.000476	0.00049	.000503	.000512	0.00052	.000535	.000545	.000558	.000572	.000588	.000604	0.00062
0	0	0	0	0	0	0	0	0	0	0	0	19187 1	38165 2	57169 4	76088 5	95348	114150	133370 9	152380 0	171360 0	190890 0	209930 0	228860 0	248250 0	267400 0	286630 0	305660 0	325070 0	343830 0	363090 0	382350 0	401910 0	421200 0	440750 0	460000	479190 0	498560 0	517810 0	537000 0	556550 0	576100 0	595270 0	614930 0	634310 0	653780	673150 0	692790	712480 0	731840 0	750350	767150 0	766850 0	766610 0	765920 0	765030 0	764160 0	000022
0	0	0	0	0	0	0	0	0	0	0	0	6521.8	9974.7	15434	20634	25142	29709	35998	42452	48546	53861	58618	63143	67907	73457	79681	85422	92005	97815	102390	106590	111370	116090	120920	125930	130930	135670	139880	143710	147550	151730	156900	162730	168960	175560	181810	187230	191960	195480	198540	204490	208290	213030	218530	224300	230320	0000000
0	0	0	0	0	0	0	0	0	0	0	0	24E-13	52E-10	19E-08	51E-07	.44E-07	51E-06	.74E-06	15E-05	42E-05	66E-05	8.7E-05	.000146	000234	0.00034	.000491	.001034	.002841	.007015	.014617	025621	.041831	.066446	0.11317	0.19844	0.3273	0.48529	0.6497	0.90304	1.4138	2.34	3.6473	5.3688	7.2263	9.1849	11.268	14.117	33.939	159.24	1148.3	3358.6	4648.9	6203.1	8128	10365	12387	
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0	0	0	0	0	0	0	0	0	0	0	0	002023	0.00446	005186	006095	007967	010011	012736	014873	016333	021587	023476	021298	024367	033063	041191	0.03657	044209	0.04795	049855	051305	050444	051039	048275	054305	054294	0.05513	059179	063498	061672	067572	066659	072046	067504	074121	073903	076907	080529	091002	096269	0.10833	093107	080615	0.06691	062804	061551	
0.75276	0.74216	0.73222	0.72196	0.7117	0.70182	0.69154	0.68123	0.67114	0.66062	0.65065	0.64029	0.62428 0.	0.61323 (0.60488 0.	0.59677 0.	0.58806 0.	0.57981 0.	0.5708 0.	0.56274 0.	0.55605 0.	0.5488 0.	0.54232 0.	0.53688 0.	0.53097 0.	0.52391 0.	0.51614 0.	0.51059 (0.50431 0.	0.49805	0.49229 0.	0.48658 0.	0.48096 0.	0.47606 0.	0.4715 0.	0.46711 0.	0.46249 0.	0.45747	0.45233 0.	0.44733 0.	0.44264 0.	0.43841 0.	0.43493 0.	0.43179 0.	0.4288 0.	0.42515 0.	0.42128 0.	0.41726 0.	0.4133 0.	0.40998 0.	0.4066 0.	0.4	0.39697 0.	0.39398 0.	0.39109	0.38852 0.	0.386 0	
0.24724	0.25784	0.26778	0.27804	0.2883	0.29818	0.30846	0.31877	0.32886	0.33938	0.34935	0.35971	0.3737	0.38231	0.38993	0.39713	0.40398	0.41018	0.41647	0.42239	0.42761	0.42961	0.4342	0.44182	0.44466	0.44303	0.44267	0.45284	0.45148	0.454	0.45786	0.46211	0.46859	0.4729	0.48023	0.47858	0.48322	0.4874	0.4885	0.48918	0.49569	0.49402	0.49841	0.49616	0.5037	0.50072	0.50481	0.50584	0.50617	0.49901	0.49713	0.49167	0.50992	0.5254	0.542	0.54868	0.55245	
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0.99409	0.99489	0.99526	0.99558	0.99585	0.99606	0.99623	0.99637	0.9966	0.99662	0.99651	0.99425	0.88777	0.8241	0.7758	0.72662	0.69205	0.65762	0.62053	0.58392	0.55559	0.52222	0.4947	0.46919	0.44575	0.439	0.46121	0.47672	0.47835	0.48498	0.49616	0.48219	0.44929	0.414	0.38043	0.37944	0.39133	0.39383	0.3684	0.33002	0.29505	0.26684	0.24787	0.23102	0.23899	0.30991	0.32173	0.31885	0.3322	0.25692 0	0.24261 0	0.23405	0.23742 0	0.26822 0	0.30356 0	0.34114 0	0.38631 0	
.005908	.005107	0.00474	.004423	.004153	.003944	.003767	.003628	.003402	.003383	.003488	005752	0.11223	0.1759	0.2242	0.27338	0.30795	0.34238	0.37947	0.41608	0.4441	0.47778	0.5053	0.53081	0.55425	0.561	0.53879	0.52328	0.52165	0.51502	0.50384	0.51781	0.55071	0.586	0.61957	0.62056	0.60867	0.60617	0.6316	0.66998	0.70495	0.73316	0.75213	0.76898	0.76101	0.69009	0.67827	0.68115	0.6678	0.73266	0.74474	0.75165	0.74509	0.71165	0.67332	0.63781	0.5967	
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31299	21531	21433	21552	21694	22081	22337	22589	21004	21820	21472	21315	19902	18116	18647	18055	17259	16716	16562	16559	15935	15608	14913	14294	13669	13862	14755	15751	16551	16987	17506	17243	16201	15201	14293	14428	15288	15497	14622	13243	11999	11073	10572	10111	10529	13715	14871	15330	17427	13009	12282	12282	12888	14693	17394	21351	26019	
186.02	110.52	102.08	95.753	90.463	87.437	84.453	82.253	71.69	74.074	75.164	123.31	2515.8	3867	5388.8	6792.9	7680.1	8703	10128	11799	12746	14279	15233	16171	16996	17713	17236	17289	18049	18039	17771	18517	19857	21517	23278	23596	23779	23853	25068	26884	28669	30424	32078	33658	33525	30540	31351	32750	35032	37097	37702	39444	40445	38984	38582	39919	40188	
27.495	27.516	27.674	27.945	28.186	28.256	28.44	28.479	12.655	25.295	27.383	25.315	11.842	12.55	13.831	14.159	14.34	14.454	14.328	14.22	14.488	14.44	14.371	14.468	14.559	14.418	14.506	14.64	14.467	14.549	14.711	14.588	14.698	14.635	14.592	14.539	14.599	14.598	14.677	14.644	14.542	14.58	14.67	14.513	14.331	14.56	13.962	12.554	7.082	7.589	7.5944	14.495	14.407	14.544	14.668	16.984	18.979	
18.31	28.568	30.569	30.112	30.364	29.96	30.031	29.949	14.141	27.306	30.078	27.938	11.214	14.833	15.429	14.057	14.878	16.083	17.34	14.975	15.422	15.581	14.656	14.777	15.396	15.895	16.009	15.57	15.295	15.435	14.622	14.769	14.972	15.041	14.804	15.429	15.262	14.858	15.119	14.713	14.593	14.654	14.972	14.627	14.868	15.088	14.112	12.493	6.5802	7.4737	7.568	14.505	13.756	13.617	13.455	14.794	15.876	
31485	21641	21535	21648	21784	22168	22422	22671	21076	21894	21547	21438	22418	21983	24035	24847	24939	25419	26690	28358	28681	29887	30146	30465	30665	31575	31991	33040	34599	35027	35282	35760	36058	36718	37570	38024	39067	39350	39690	40127	40669	41498	42650	43769	44054	44256	46222	48080	52459	50634	50624	52477	54283	54779	57300	62588	67351	
20967	22469	23788	23327	23468	23505	23676	23842	23551	23635	23668	23659	21229	25983	26812	24669	25876	28284	32299	29863	30530	32249	30743	31115	32429	34809	35307	35139	36579	37158	35069	36204	36729	37737	38116	40350	40841	40049	40884	40315	40811	41708	43529	44114	45706	45860	46719	47846	48741	49864	50448	52514	51831	51291	52561	54518	56338	
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2787.3	2731	2769.2	2809.1	2842.1	2871.9	2897.2	2920.1	2959.5	2993.9	3032.6	3073.8	3128.9	3152.7	3234	3256.6	3273.6	3315.2	3354.7	3405.2	3420.4	3461.3	3477	3487.5	3504.1	3544.3	3592.7	3624.6	3663.8	3689.3	3699.8	3705.3	3710	3712.5	3725.1	3743.1	3761.3	3773.7	3782	3787.2	3790.9	3795.5	3809	3813.8	3820.7	3837.8	3842.7	3837.3	3806.8	3791.5	3788.6	3793	3787.2	3762.7	3728.9	3676.6	3619.1	
37.777	69.23	98.229	127.95	157.54	185.91	215.09	244.04	272.76	302.66	331	360.45	402.28	431.69	456.48	482.93	509.87	534.47	558.71	580.79	603.88	625.74	647.76	669.65	691.5	711.6	731.2	750.53	769.32	787.62	806.75	825.77	844.92	863.45	881.52	898.88	915.87	932.97	950.01	966.9	983.92	1000.6	1016.2	1032.1	1047.2	1062.3	1077.2	1091.9	1106.2	1120	1133.6	1159.8	1172.6	1185.8	1198.9	1211.9	1224.1	
0.010058	0.020188	0.030063	0.040261	0.050365	0.060075	0.070122	0.080152	0.090121	0.10038	0.11016	0.12032	0.13478	0.14477	0.15477	0.16474	0.17486	0.18473	0.19476	0.20475	0.21466	0.22486	0.23486	0.24473	0.25483	0.26477	0.27486	0.28469	0.29482	0.30467	0.31471	0.32467	0.33481	0.34471	0.3548	0.36474	0.37467	0.38472	0.39472	0.40466	0.41476	0.42484	0.43474	0.44484	0.45471	0.46476	0.47468	0.48471	0.49475	0.50467	0.51468	0.53434	0.54414	0.55421	0.56427	0.57429	0.58415	



0.000635	0.000646	0.000655	0.000661	0.000667	0.000672	0.000676	0.00068	0.000683	0.000686	0.000689	0.000691	0.000693	0.000692	0.000699	0.000706	0.000712	0.000718	0.000722	0.000726	0.000729	
763340	761950	759040	755590	752420	749660	747210	745020	743000	740960	739100	737270	735610	746860	744650	742280	740670	739550	738740	738020	737360	
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15584	17602	20743	24326	27652	30534	33116	35424	37594	39741	41693	43596	45336	46323	48643	51213	52982	54254	55160	55974	56708	
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0.055274	0.054163	0.053544	0.053001	0.052334	0.051884	0.05145	0.050849	0.050513	0.049847	0.049543	0.04923	0.048934	0.047104	0.046646	0.046131	0.045703	0.045317	0.045053	0.044777	0.044585	
0.38079	0.37827	0.37604	0.37388	0.37173	0.36958	0.36747	0.36539	0.36332	0.36127	0.35933	0.35735	0.35546	0.35139	0.34244	0.33538	0.32964	0.32471	0.32044	0.31676	0.31346	
0.56394	0.56757	0.57042	0.57312	0.57594	0.57854	0.58108	0.58376	0.58617	0.58888	0.59113	0.59342	0.5956	0.60151	0.61091	0.61849	0.62465	0.62997	0.63451	0.63846	0.64196	
0.01931	0.026663	0.045567	0.042115	0.037227	0.03261	0.02917	0.027259	0.025313	0.024825	0.023333	0.022059	0.021053	1.16E-23	0.007888	0.004788	0.003622	0.002461	0.001937	0.001517	0.001226	
0.32291	0.2366	0.22718	0.22438	0.22292	0.22084	0.21996	0.21749	0.212	0.20832	0.20337	0.19927	0.19627	0.17368	0.15624	0.12071	0.10191	0.093166	0.077534	0.068765	0.061577	
0.65778	0.73674	0.72726	0.73351	0.73985	0.74655	0.75087	0.75525	0.76269	0.76685	0.77329	0.77867	0.78267	0.82632	0.83587	0.87451	0.89447	0.90437	0.92053	0.92972	0.9372	
1306.2	1717.4	2944.3	2728.5	2424.3	2137	1927.5	1808.2	1698.3	1669.3	1574.9	1493.5	1431.1	2.89E-19	199.36	121.37	91.628	62.139	48.832	38.248	30.902	
21844	15239	14679	14537	14517	14472	14535	14427	14223	14008	13727	13492	13342	4330.7	3949	3059.4	2577.9	2352.7	1954.6	1734.4	1552.7	
44496	47454	46991	47522	48181	48921	49615	50100	51170	51565	52196	52720	53204	20604	21126	22165	22627	22838	23206	23449	23631	
23.15	23.698	24.902	25.447	25.723	25.41	25.467	25.598	26.011	26.132	26.101	26.051	26.108	26.574	26.564	26.74	26.755	26.835	27.019	26.925	26.911	
21.237	23.287	24.603	25.237	25.533	25.195	25.204	25.425	25.706	25.922	25.919	25.896	25.945	26.812	26.443	26.542	26.61	26.736	26.965	26.858	26.852	
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62058	63295	63837	64252	64640	64977	65394	65888	66307	66702	67028	67305	67554	25159	25159	25159	25159	25159	25159	25159	25159	
26038000	26255000	26437000	26616000	26795000	26968000	27138000	27305000	27470000	27634000	27787000	27943000	28089000	28949000	29552000	30045000	30446000	30792000	31087000	31342000	31570000	
48.463	48.865	49.205	49.538	49.87	50.194	50.51	50.821	51.128	51.432	51.718	52.007	52.28	53.88	55.002	55.92	56.667	57.311	57.86	58.334	58.758	
3484.2	3459.4	3454.6	3451.3	3445.9	3442.8	3439.2	3435.3	3429.7	3425.2	3421.8	3418.6	3415.6	2692.1	2689.8	2675.9	2664.8	2654.2	2647.1	2640.7	2634.9	
1247.4	1258.3	1269.2	1279.7	1290.4	1301	1311.4	1321.8	1332.3	1342.8	1352.8	1363.2	1373.2	1509.7	1646.5	1782.7	1918.7	2055.7	2191.5	2327.3	2463.9	
0.60424	0.61422	0.62435	0.6342	0.64421	0.65423	0.66416	0.67413	0.6842	0.69438	0.70421	0.71442	0.72424	0.77449	0.82468	0.8747	0.92461	0.97488	1.0247	1.0746	1.1247	



[Appendix 4]

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