THE PROCESS OF *ENHANCED OIL RECOVERY* (EOR) USING *ALKALINE SURFACTANT (AS) FLOODING* ON SUKOWATI CRUDE OIL: A COMPARISON STUDY OF MIXED SURFACTANT

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Abstract. We investigated six mixed surfactants for effectiveness in enhanced oil recovery (EOR). The selected surfactant formulation were tested for EOR using coreflood tests beneath sandstones. There are four objectives in this research. The first objective would examine synergism of surfactant mixtures, i.e., MES-SLS; MES- LAS; MES-DEA; MES-Tween 80; MES-BAC; and BAC-Tween 80. The next study examines the optimal ratio of the best surfactant mixture. The next step is to determine the best alkali between NaOH and Na2CO3. The final objective would determine the optimal concentration of alkaline in this formulation. Surfactant formulation would use brine 5000 ppm. This formulation injected with a flow rate of 10 ml/s at temperature of 70oC for 3 minutes beneath sandstones. Oil recovery results and the IFT value are observed response in this study. This research has found MES-DEA (½: 1) and alkali sodium carbonate 1% wt/v as the best formulations. This formulation has given 32,88% wt oil recovery and IFT value of 3,29 x 10-1 mN/m.

Keywords : Alkaline, AS flooding, Enhanced Oil Recovery (EOR), mixed surfactant

INTRODUCTION

Petroleum recovery processes can be divided into three phases, there are primary, secondary and tertiary phases. Primary phase applies a natural process that depends on the natural energy resource in the reservoir. Secondary phase applies the process of immiscible gas flood and water flood. If the the residual oil at the end of secondary phase is still deserved to be obtained, so further, there will be oil obtaining at the tertiary phase (tertiary recovery) on the reservoir stones. The method on this tertiary phrase is called as Enhanced Oil Recovery (EOR) method (Green and Willhite, 1998). The oil obtaining in some reservoirs,with the primary phase (15-20%) and the secondary phase (15-25%) could bring back just a half of total number of the oil (Holmberg et al., 2002).

The EOR method involves the injection process of the material which can cause changes in the reservoir like the oil composition, mobility ratio, and the characteristic of fluid-stones interaction. Based on the material which is injected into the EOR reservoir, it can be distinguised to some methods, they are; the heat method (hot water, steam stimulation, steam flood, fire flood), chemical method (polymers, surfactants, alkaline), solvent-miscible method (hydrocarbon solvent, CO2, N2, hydrocarbon gas, mixed of natural gases) and the others (foam, microbial) (Green and Willhite, 1998).

With EOR method, the oil production can enhance as much as 30-60% (US Department of Energy, 2011). Even be more than it, compared by primary and secondary recovery which only totally extract the oil as much as 20-40 % from the reservoir (Electric Power Research Institute, 1999).

The chemical flood is done by injecting some chemicals such as surfactants, polymers, and alkaline, either by individualy, combined or sustainable on some old wells that are considered as potential wells. The using of surfactants is to decrease oil-water interfacial tension (IFT) in the reservoir, so that it will increase the displacement efficiency (Flaaten et al., 2008). Although there are many EOR techniques which can be choosen and have been tested on some cases, but the using of surfactant is the most recomended because of its ability to decrease the IFT value to the lowest (Zhang et al., 2007).

Research about chemical EOR has been studied by some countries for a long time since 80s. recent research with various surfactant formulations has been applied with all reviews. For examples: the use of mixed surfactants with two group functions (Barger et al., 2006); the use and ability of sulfate anionic surfactant (Liu et al., 2007; Dong et al., 2008; Iglauer et al., 2010). The performance of sulfonate anionic surfactant (Zhang et al., 2007; Campbell & Sinquin, 2008; Sandersen et al., 2012; Zhou et al., 2013). The performance nonionic surfactant (Li et al., 2009; Iglauer et al., 2010; Dong et al., 2013; Park et al., 2014); review about the influences of the kinds of surfactans (anionic, cationic and nonionic) toward kinds of reservoir (Hou et al., 2015; Wei et al., 2015); a research about surfactant with various kins of lenghts of chains and its combination with alkali (Sharma et al., 2015).

Research about surfactant formulation is also done by mixing the primary surfactant and the secondary surfactant together with the solvent and it is used when the core flooding, and then the result can enhance the oil earning as much as 85% (Campbell et al., 2009).

Another research done by mixing strong anionic and the weak one and thenn added to solvent, co-surfactant, polimer and alkali. It is found that the surfactant will be more dissolved in the brine. This result of formulation is able to fulfill the requirement of the parameter of screening test surfactant EOR, that is IFT value reach the number of 10^{-3} dyne/cm (Berger et al., 2007).

In a simple research, some kinds of surfactants are well-mixed to improve the characteristics of the product at the end process. That mixing aims to get the synergism of surfactant mixture. It also aims to get a better characteristic of the mixture than the characteristic of invidual component. There are two main problems on the design of surfactant formulation: first, the inter facial tension is enough for mobilizing oil that is trapped; second, the surfactant must keep the integrity during the movement through the reservoir. With alkali surfactant injection method, it is able to decrease the inter facial tension as being expected.

METHODOLOGY

MATERIALS AND TOOLS

The materials used in this research are crude oil from Sukowati refinery (East Java), sulfonate anionic surfactant: (linear alkilbenzene sulfonate (LAS) (code S00505); metyl ester sulfonate (MES); sulfate aninonic surfactant : (sodium lauryl sulfate (SLS)), cationic surfactant: (benzalconium chloride (BAC P109.50), and nonionic surfactant: (tween 80 TW 0-120; diethanolamide (DEA)) were used. Natrium chloride was prepared with concentration 5.000 ppm. Kali Babon sandstone was used for modeling. All the solutions were prepared in distilled water.

The tools used in this research are waterbath thermostat, submerged pump, control valve, venturimeter, thermometer, picnometer, Spinning Drop Interfacial Tensiometer TX500D, tensiometer KRUSS, viscosimeter brookfield RVDVE serial no. E6509104, centrifuge, heater were used. The scheme of experimental tools in the study presented in Fig. 1.

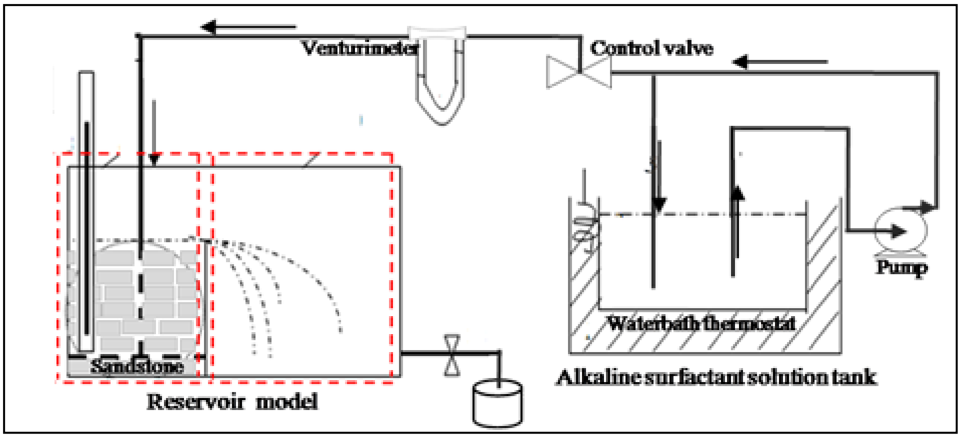

Fig 1. The Series of Experimental Tool.

EXPERIMENTAL METHODOLOGY

Waterflood

Sandstones are arranged on a model of reservoir together with the oil resource with some variations of oil- stone ratio1:5, 1:10, 1;15, 1:20, 1:25. Inject the water and brine solution suitable with levels of oil resources for 2 minutes with the rate of flow 1 ml/second into the reservoir at the temperature of 70^{o}C. The emulsion of overflow is stored in the tank and then measure the oil recovery that is obtained after 24 hours until oil and water is well-separated.The experiment is conducted to obtain the proper volume ratio oil-stone which will be applied for EOR main research.

Compatibility Test

The compatibility test is a test of compatibility of the brine water conducted by dissolving each surfactant (MES, LAS, SLS, DEA, Tween 80, dan BAC) into the brine water with concentration of 0,16 M Then, this surfactant solution is combined with volume ratio 1:1 and added with alkali for each combination. After that, just let it stuck on it's place and then oberved whether the surfactants will dissolve in synteic-formed water or not. If during the storage, there is no indication of lumps or sediment, it means that the mixing of surfactans solution and alkali is stable.

Surface Tension Measurement

Surface tension is measured using tensiometer Kruss (Germany) at temperature 70^{o}C with Ring method.

Interfacial Tension Measurement

Interfacial tension is measured using Spinning Drop Interfacial Tensiometer TX500D at temperature 70^{o}C and rotation of 6.000 rpm.

Core Flooding

Sandstones and sample of oil resources are arranged on a model of reservoir with oil-stone ratio based on variables obtained at water flooding experiment. The injection of oil into the stone is done for 24 hours. The surfactant solution that has been formulated based on the set temperature and condition then will be injected into the the model of reservoir until being overflow for about 3 minutes with the flow rate of 10 ml/second. After 24 hours, the oil that has been out with an injectable solution is separated to find out oil yield which has been recovery.

RESULT AND DISCUSSION

Characterization of Crude Oil

The results of the characteristics of the crude oil used shows in Table 1.

Table 1. Characterization of Crude Oil

Characterization	Result of Observation
Physical form	Dark brown viscous oil, distinctive odor of crude oil.
Density (*SpesificGravity*,g/ml)	0,8181 (30°C); 0,7980 (70°C)
Viscosity (mPas)	52,8(30°C); 2 (70°C)
% Aspalten	6,15%b

The density dan viscosity of Sukowati crude oil decreases significantly along with ine increasing of temperature. At a room temperature, the oil will curdle so that the surfactant formulation that is suitable with its the characteristic, later will determines the result of oil recovery.

The number of viscosity can indicate the movement of droplet in the emulsion. If the viscosity value is high, so the droplet inside the emulsion will be hard to move. It is also influenced by aspalten contents- a natural emulsifier inside the crude oil. The content of aspalten in Sukowati crude oil is 6,15%b. The aspalten content in the oil plays an important role in decreasing inter facial tension if it is added by an alkali, IFT aspalten solution with alkali is lower than IFT of crude oil and low aspalten and alkali solution (Guo et al., 2006).

Water Flooding

Water flooding test is used to determine the ratio of oil- rock where the brine injection can not afford oil recovery. Rock porosity is only 9,80%, so the ability of oil retained only for a maximum of the porosity. Optimum ratio of oil-rock was found by 1:15. In this study used this ratio to EOR modeling.

The Result of Compatibility Test

Surfactant solution is said to meet the compatibility test as a chemical surfactant flooding is that if the solution to form an emulsion O/W, clear solution, not cloudy (hazy solution) and does not form a precipitate. The test results are presented in Table 2. All the surfactant formulation meets this test and can proceed to the next test.

Table 2. Compatibility Tests Result

Mixed Surfactant	Compatibility
MES-LAS	Light milky, rather cloudy, soluble
MES-SLS	Light milky, rather cloudy, soluble
MES-DEA	Light milky, rather cloudy, soluble
MES- Tween 80	Light milky, rather cloudy, soluble
MES-BAC	Clear, soluble
BAC– Tween 80	Clear, soluble

Surface Tension Measurement Result

Surface tension measurement results shown on Table 3. Alkali surfactant formulations produce SFT value varies from the smallest of 25,33 mN/m and 30,57 mN/m for the biggest, each other for mixture MES-DEA and MES-Tween 80. Both of them are mixture of sulfonate anionic and nonionic surfactant.

According to research Dong et al. (2013) SFT value increases with increasing number of units (OCH_2CH_2) or have greater molecular weight, using anionic-nonionic alcohol ethoxylates propanesulfonic acid (AES-7,9,10) value of γcmc 34,19; 41.96; 44,50 mN/m and nonylphenol propanesulfonic acid (NPS-4,9,10) value of γcmc 31,62; 34,50; 36,15 mN/m. But in this study, it is found no correlation between molecular weight to the value of SFT. This research obtains SFT value of formulation smaller than their formulation.

Table 3. SFT Value of Surfactant Formulation

Component	γ_{cmc} (mN/m)
MES-DEA	25,33
MES-BAC	27,43
BAC-Tween 80	28,00
MES-SLS	28,43
MES-LAS	29,26
MES-Tween 80	30,57

Interfacial Tension Measurement Result

Based on the measurement results surfactant formulations IFT, the results showed that the formulation of the interface tension values ranging from 10^{-1} mNm^{-1}. Measurement of interfacial tension using a spinning drop tensiometer based on the principles of geometry decline, where the influence of the difference in density between the two liquids and rotation determine IFT values obtained (Pojman et al., 2001).

The smallest IFT values obtained in the formulation MES-DEA with a volume ratio of 1:

2 and alkali (Na_2CO_3) 1%, as much as 3,29 x 10^{-1} mNm^{-1}. The largest IFT in the formulation MES-BAC with a volume ratio of 1: 1 and alkali (Na_2CO_3) 0.5%, amounting to 6,57x$10^{-1}$$mNm^{-1}$. Interfacial tension measurement result shown on Table 4. The form of oil droplets in the surfactant solution when measuring the IFT value are show in Fig.2, 2a to surfactant MES-DEA and 2b for MES-BAC. A low IFT value is achieved when the diameter droplets is small.

Table 4. IFT Value of Surfactant Formulation

Surfactant Formulation	IFT (mN/m)
1. MES-DEA (1:1) 0,5% Na_2CO_3	4,52 x 10^{-1}
2. MES-DEA (½:1) 0,5% Na_2CO_3	4,13 x 10^{-1}
3. MES-DEA (2:1) 0,5% Na_2CO_3	4,58 x 10^{-1}
4. MES-DEA (½:1) 1,0 % Na_2CO_3	3,29 x 10^{-1}
5. MES-BAC (1:1) 0,5% Na_2CO_3	6,57 x 10^{-1}

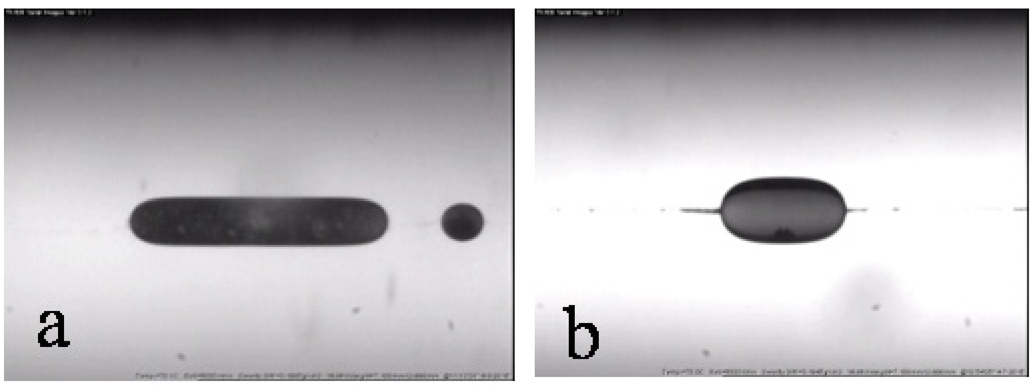

Fig 2. Shape of Droplets in IFT Measurement (a) drop width MES-DEA, (b) drop width MES-BAC.

Alkali surfactant mixture is more effective in lowering the interfacial tension at EOR. Fig. 3 illustrates the dynamic measurement of IFT of various alkali surfactant mixtures during the measurement process. Within the same measurements, it is obtained an IFT values which is relatively stable. The stability of the IFT in a achieving ultra low IFT because of the alkaline that is combination with a surfactant.

Dong et al. (2013) ever examine the dynamics of IFT measurements between anionic-nonionic surfactant solution in comparison with the addition of alkali-surfactant of each on crude oil with acid number 11,51 mgKOH/g crude oil (density 0,918 g mL^{-1} at 25 °C), it is obtained the low IFT in a short time (<10^{-2} mN/m, within 3 minutes) by a mixture of alkali only for the surfactant solution itself while for the surfactant solution requires a longer time (100 mN/m, within 30 minutes). The addition of alkali is very important to lowering the interfacial tension at EOR research.

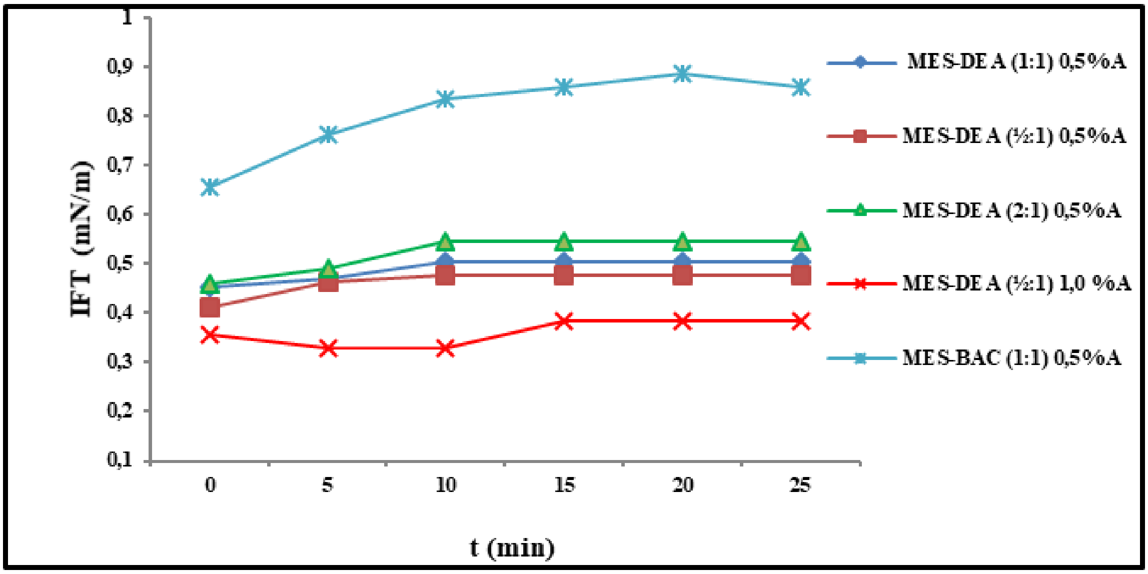

Fig. 3. Curve of IFT Measurement of various Alkaline Surfactant Mixture

The Determination of Best Surfactant Formulations

Oil yields which is obtained from a mixture of surfactants is presented in Fig. 4, It shows the highest recovery by MES-DEA mixture as much as 25,88%. The lowest result by a mixture of surfactant MES-BAC (anionic-cationic) is 12,74%.

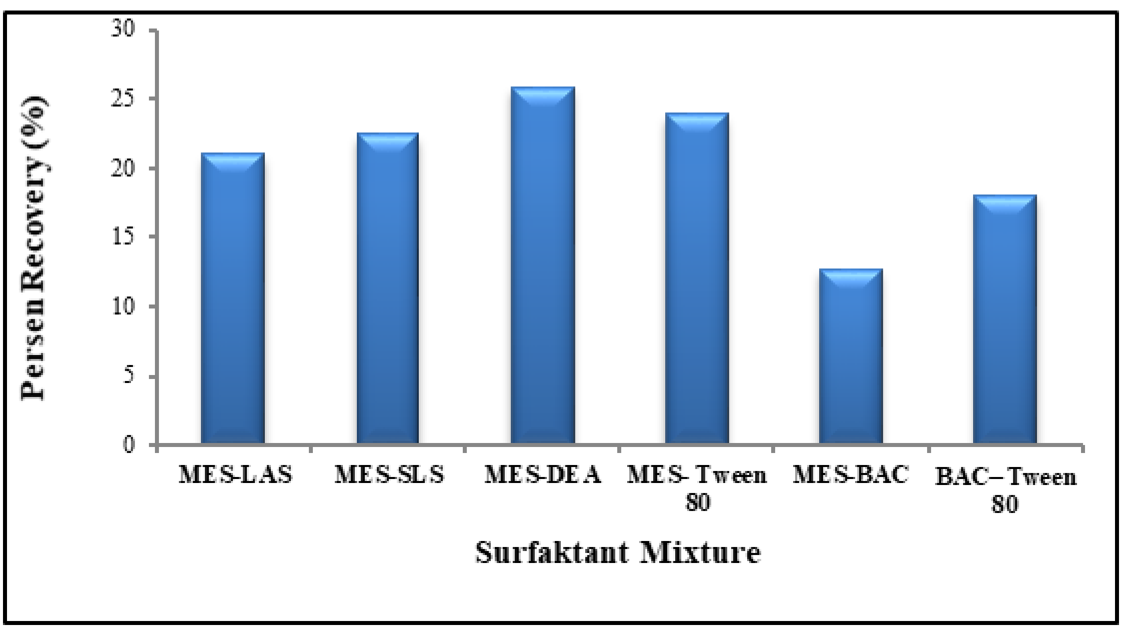

Fig. 4. Results of Recovery Surfactant Formulations

Based on the experimental results of modeling test EOR, it is found that a mixture of surfactant MES-DEA has the ability to lift the greatest oil surfactant mixture among others. MES is a sulfonate anionic surfactant containing hydrophobic greater than its hydrophilic, so it is less polar and less soluble in water and tends to form two phases. DEA is a nonionic surfactant having a hydrophilic larger content. So that a mixture of both at appropriate concentrations can improve the solubility of one another and meet the compatibility test. This mixture also gives the lowest value of SFT and IFT compared to other mixtures. Iglauer et al. (2010) conducted a surfactant formulations using DEA 2 wt.% with the solvent 2-propanol 1%wt in brine 40.000-100.000 ppm get IFT value $2x10^{-3}$ mN/m dan and yield recovery of oil by 75%. This result is the smallest IFT value in his research. Other results states that the absorption of surfactants DEA is also the smallest in the kaolinite as much as 17mg/g (Iglauer et al., 2010). Research Yin and Zhang (2013) which measures the IFT value of some types of surfactants,they found IFT lowest value of $1,1 \times 10^{-3}$ mN/m by 0,5 wt.% nonionic surfactant than other types of surfactants.

This result is also linear with previous research that gets results mixing anionic surfactant (SDBS) and nonionic (TX-100) were able to reduce the electrostatic repulsion, lowering value the CMC, and improve the interaction between them and brine, and leads to the formation of micelles which is small so that the effect of mixing is better (Wei et al., 2015). Research derivative of petroleum sulfonate surfactant mixing with nonionic surfactants are also carried out by Hongyan et al. (2009) and get IFT value $2,95 \times 10^{-3}$ mN/m with 18,1% OOIP oil recovery.

MES-DEA is the best combinations in this study because of each had a good performance, and the incorporation improve the characteristic of each. Surfactant mixture MES-BAC has the lowest ability in lifting Sukowati crude oil that is as much as 12,74%. This mixture is a mixture of anionic and cationic surfactants. Is actually a mixture of the two surfactants which is not compatible (Kronberg et al., 2014). However, BAC surfactants here have shorter chain of MES so that mixing provides enough good compatibility in the initial screening test surfactant.

Determination of Best Surfactant Mixture Ratio.

Test on a ratio comparison on the best surfactant mixture is a mixture of surfactant MES-DEA, conducted with volume ratio ½: 1. It is obtained the recovery result as shown in Fig. 5.

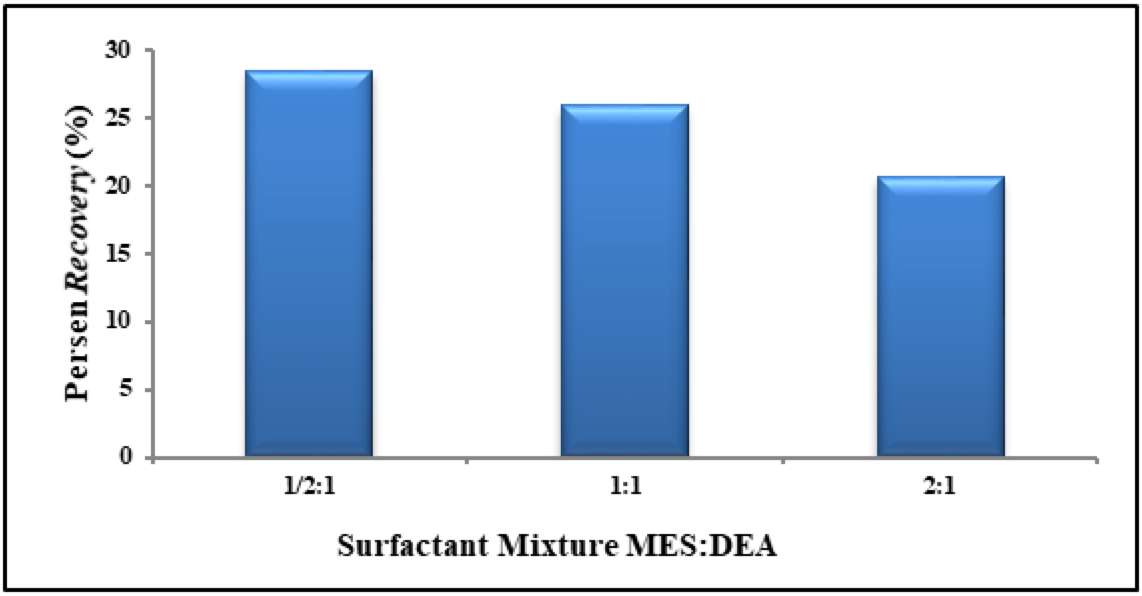

Fig. 5. **Recovery Surfactant Formulation MES-DEA With Ratio Variable**

The greater the concentration of DEA in the solution, the ability of the resulting surfactant formulation is getting better. This is because the greater content of DEA will help solubility of MES and then the solution will be more compatible.

Based on IFT test It was also obtained relevant results with the results of its recovery. IFT value MES:DEA ½:1, 1:1 and 2:1 sequently are 4,13 x 10^{-1}; 4,52 x 10^{-1} and 4,58 x 10^{-1} mN/m. The measurement result shows that the greater part of the DEA, the better in improving the characteristic of the mixture by lowering the interfacial tension.

Determination of Best Alkali Selection.

The test in selecting alkali between NaOH with Na_2CO_3 is conductedusing a combination of surfactant MES-DEA with ratio of ½: 1 with an alkali concentration of 0,5 wt.%. In this study, It is obtained that recovery of Na_2CO_3 28,52% is better than 12,26% NaOH. This is because natrium carbonate can reduce the rate of ion exchange and release of mineral (beneath sandstone) better compared with natrium hydroxide, moreover, it can also reduce the adsorption of anionic surfactant in the reservoir stone and suppress the concentration of calcium ion (Ca^{2+}). Carbonate sediment does not affect the permeability if it is compared with hydroxide and silicate, thus result of recovery with this alkali is also higher (Hirasaki and Zhang, 2003). Na_2CO_3-surfactant synergism is also reported by Qiao et al. (2012), it is providing a very low IFT values.

Determination of Best Alkali Concentration.

Natrium carbonate is considered as the better alkali than NaOH in previous studies. Alkali concentration varied from 0,25; 0,5; 0,75 and 1,0%. Concentration used up to 1,0%, the concentration used is good enough, because the higher concentration of alkali, the surfactant solution in the brine water is more turbid, in addition when it is applied in large amounts, it would be more economical. Results of research of determining the alkali concentration showed that there is an increasing recovery in percent along with increasing concentration of alkali as shown in Fig. 6.

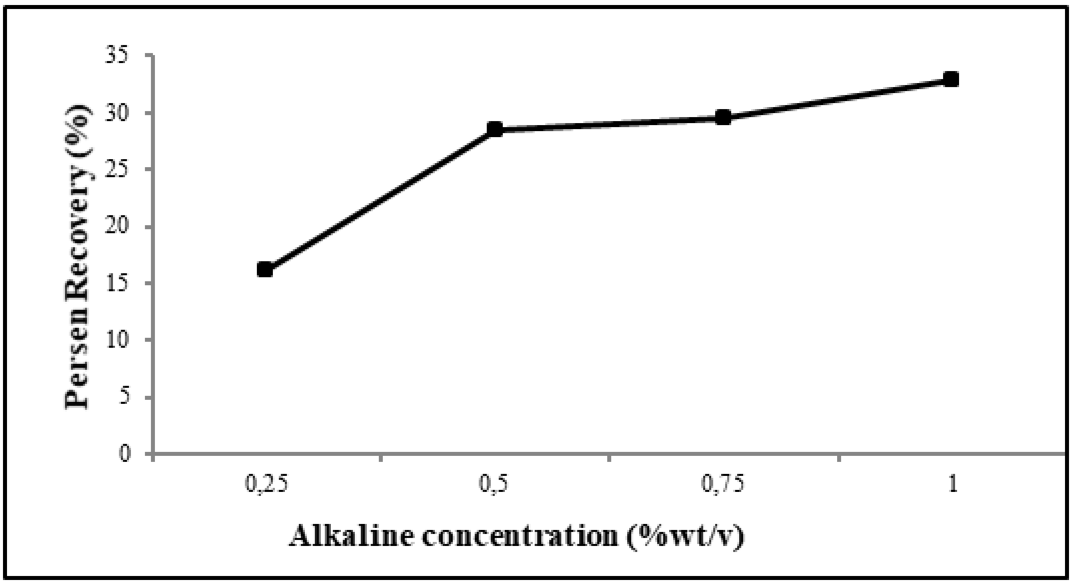

Fig. 6. The Relation Between Alkali Concentration and Percent Recovery

At the highest concentration of 1%, it is obtained the percent recovery of 32,88%. The higher the concentration of alkali, the grater oil that is obtained, but the increase of percent recovery of a concentration of 0.5% w/v to 1% w / v is relatively insignificant. Sedaghat et al. (2013) conducted a study the effect of alkali (Na_2CO_3) concentration and it showed that oil recovery yield increases with the increase the concentration of alkali. Khan et al. (2009) showed that the use of other alkali solution, it was decreasing IFT value with increasing concentrations of alkali to a concentration of 1%, when the alkali concentration is above 1%, the IFT value produced relatively constant.

CONCLUSIONS

Ratio of an optimal stone oil in a model of EOR is 1:15, in accordance with the magnitude of stone porosity. The order of Surfactant mixture which gives percent recovery of from the biggest is MES-DEA, MESTween 80, MES-SLS, mes- LAS, BAC-Tween 80 and MES-BAC. The best ratio of surfactant mixture is the ratio of MES: DEA ½: 1, 1: 1 and then 2: 1. The addition of Na_2CO_3 alkali obtains a better recovery results than NaOH. The higher the concentration of alkali used, the higher the percent of recovery and the lower IFT value of 3,29 x 10^{-1} mN/m (at a concentration of 1% alkali).

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