SURFACTANT SELECTION FOR ENHANCING EMULSION MOBILITY USING INTERFACIAL TENSION MEASUREMENT

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การคัดเลือกสารลดแรงตึงผิวเพื่อเพิ่มการเคลื่อนที่ของอิมัลชันโดยการวัดแรงตึงระหว่างผิว

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วิทยานิพนธ์นี้เป็นส่วนหนึ่งของการศึกษาตามหลักสูตรปริญญาวิศวกรรมศาสตรมหาบัณฑิต
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การเคลื่อนที่ของอิมัลชันเป็นลักษณะสำคัญของของไหลที่ช่วยเพิ่มการผลิตน้ำมัน ในงานวิจัยนี้สารลดแรงตึงผิวถูกนำมาใช้เพื่อช่วยเพิ่มการเคลื่อนที่ของอิมัลชัน โดยหน้าที่หลักของสารนี้คือจะช่วยลดแรงตึงระหว่างผิวของของไหลทั้งสองชนิดและจะทำให้อิมัลชันเคลื่อนที่ได้ง่ายในแหล่งกักเก็บ วัตถุประสงค์ของงานวิจัยนี้คือทำการวัดค่าแรงตึงระหว่างผิวภายใต้เงื่อนไขแหล่งกักเก็บน้ำมันในประเทศไทย และทำการวิเคราะห์ผลกระทบของพารามิเตอร์ต่างๆในแหล่งกักเก็บที่อาจจะส่งผลต่อการลดค่าแรงตึงระหว่างผิว เช่น ความดัน อุณหภูมิ ค่าความเค็ม ชนิดของสารลดแรงตึงผิว และค่าความเข้มข้นของสารลดแรงตึงผิว

ผลการทดลองพบว่า ความดันในช่วง 1,000 ถึง 2,000 ปอนด์ต่อตารางนิ้ว ณ อุณหภูมิคงที่มีผลกระทบต่อแรงตึงผิวน้อย สำหรับผลกระทบจากชนิดของสารลดแรงตึงผิวที่ระดับความเข้มข้นที่เท่ากัน สารที่ให้ประสิทธิภาพสูงที่สุดคือ สารโมโนเอทาโนลามีน สารดังกล่าวมีความเป็นเบสเนื่องจากในส่วนหัวของโมเลกุลมีกลุ่มของแอมีน จึงทำให้มีความสามารถในการผลิตสารลดแรงตึงผิวภายในได้มากขึ้น ความเข้มข้นของสารลดแรงตึงผิวมีบทบาทสำคัญในการลดแรงตึงระหว่างผิวโดยมีผลมากถึง 87.13% ส่วนการเพิ่มอุณหภูมิจาก 70 ถึง 90 องศาเซลเซียส จะมีผลต่อการลดแรงตึงผิวเพียงเล็กน้อย นอกจากนี้แรงตึงผิวจะลดลงเมื่อเพิ่มความเค็มของสารละลาย อย่างไรก็ตามหากความเค็มสูงเกินไปจะไม่สามารถช่วยลดแรงตึงผิวได้ ส่วนไดวาเลนต์ไอออนส่งผลกระทบต่อค่าความตึงผิวค่อนข้างน้อย

จากการศึกษาผลกระทบของพารามิเตอร์ต่างๆเหล่านี้จะช่วยเพิ่มความเข้าใจกระบวนการในการลดค่าความตึงระหว่างผิว และจะเป็นข้อมูลพื้นฐานในการนำไปใช้ในอนาคตสำหรับการเพิ่มการผลิตน้ำมัน

ภาควิชา วิศวกรรมเหมืองแร่และปิโตรเลียม ลายมือชื่อนิสิต ....

สาขาวิชา วิศวกรรมทรัพยากรธรณีและปิโตรเลียม ลายมือชื่อ อ.ที่ปรึกษาหลัก ....

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JIRAMET JIRAVIVITPANYA: SURFACTANT SELECTION FOR ENHANCING EMULSION MOBILITY USING INTERFACIAL TENSION MEASUREMENT. ADVISOR: ASST. PROF. KREANGKRAI MANEEINTR, Ph.D., 89 pp.

Emulsion mobility is the crucial characteristic of fluid to increase oil production. In this study, surfactants are considered as chemical agents used in Northern oilfield, Thailand. The important role of surfactant is to reduce the interfacial tension (IFT) of two fluids and to make emulsion flow easier in the reservoir. The aim of this work is to investigate and measure the IFT based on the conditions of subsurface at the oilfield in Thailand. These parameters such as temperature, pressure, salinity, types of surfactant as well as the concentration of surfactant are adjusted to investigate the effects on IFT reduction.

The results show that the pressure ranging from 1,000 psi to 2,000 psi at constant temperature has less significant effect on IFT reduction. For the effect of types of surfactant solution, monoethanolamine (MEA) is the best type of surfactant samples which has higher performance to reduce interfacial tension at the same concentration. Because of its amine group, MEA has a strong base that can dissociate in the solution and create more in-situ surfactant to lower IFT. Surfactant concentration is the main parameters that impact on the IFT reduction. It can greatly decrease IFT up to 87.13% for surfactant concentration. Temperature varied from 70C to 90C can insignificantly reduce the IFT. In addition, the increase of salinity can reduce interfacial tension, but it is relatively stable at high salinity. Lastly, the effect of divalent ions has less impact on the interfacial tension.

These investigated effects of each parameter will be useful to understand the mechanism of IFT reduction, and they will be used as fundamental data to apply for oil recovery with reservoir conditions.

Department:	Mining and Petroleum Engineering	Student's Signature	....
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CMC	Critical Micelle Concentration
DAS	Drop Analysis System
DEA	Diethanolamine
EOR	Enhanced Oil Recovery
FAOH	Fatty Alcohol
IFT	Interfacial Tension
MEA	Monoethanolamine
RG	Refined Glycerine
wt.%	Percent by Weight

NOMENCLATURE

m	Mass
P	Pressure
R	Radius of Curvature
V	Volume

GREEK LETTER

ρ	Density
γ	Interfacial Tension

CHAPTER 1
INTRODUCTION

1.1 Background

The world’s principal energy is fossil fuels. Every year the world economic growth is developing continuously that caused by demand and energy consumption growing rapidly (Sheng, 2011). In contrast, the energy production declined, resulting in more imports to encounter the domestic demand. Also, the resource development by using out-of-date techniques cannot produce sufficient oil, and producing unconventional resources are more expensive than conventional resources. In order to reduce the oil import in Thailand and to recover oil to meet the domestic demand, enhanced oil recovery (EOR) techniques are more concerned. Therefore, emulsion mobility needs to be concerned to serve this rising demand.

Oil recovery operations can be classified into three phases: primary recovery, secondary recovery and tertiary recovery. Primary recovery is the first stage that recovers oil by using natural drive energy existing in the reservoir such as solution gas, water influx, gravity drainage, gas cap, rock and fluid expansion. Secondary recovery is the next stage when natural drive energy is insufficient to push oil up to the surface. Nevertheless, there is unsatisfied amount of oil for primary recovery. Thereby, water is injected as secondary recovery which is used to maintain the pressure in reservoir to increase oil production. This process can be generally called waterflooding. However, disadvantage of waterflooding is lower in volumetric sweep efficiency that is defined as the ratio of oil volume contacted by the injected fluid to initial volume of oil in place. Therefore, special fluids are significantly required for injecting in the reservoir to increase oil recovery. This method is called enhanced oil recovery (EOR) technology.

1.2 Enhanced Oil Recovery Technologies

Enhanced oil recovery (EOR) technology is getting more attention because of ineffective primary and uneconomical secondary recoveries. Applying enhanced oil recovery processes can give an incremental recovery of oil as tertiary recovery. The tertiary recovery processes can also be divided into several methods such as thermal recovery, miscible flooding, microbial flooding, and chemical flooding (polymer, surfactant, alkaline etc.). Figure 1.1 shows the EOR methods used in the petroleum industry. This study will only focus on chemical flooding that is conducted as a suitable method to increase oil recovery (Lyons, 1996).

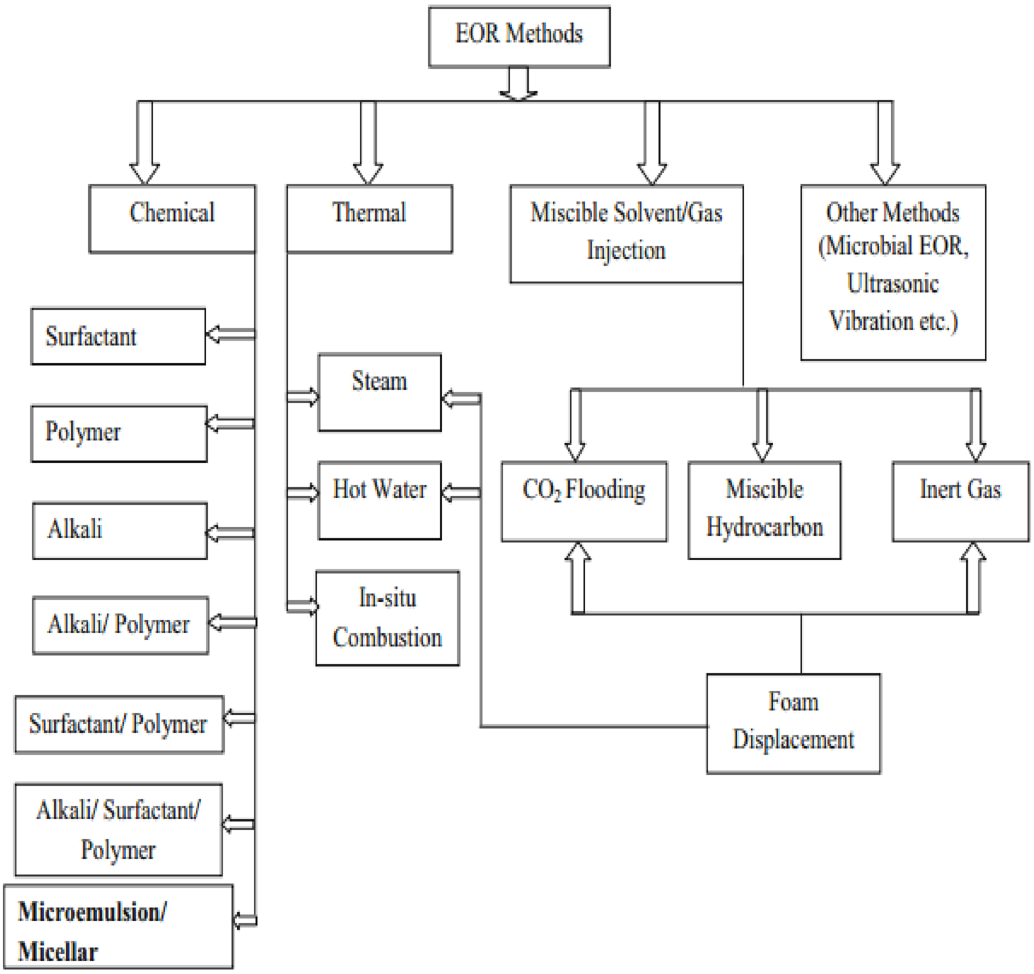

Figure 1.1 Classification of EOR processes (Bera & Mandal, 2015)

1.3 Chemical Flooding

For chemical flooding, oil recovery is improved by means of injecting fluid which is immiscible with the displaced phase. Most commonly used chemicals are polymer, surfactant, and alkaline. These types of fluid can interact with the reservoir rock and fluid system to create favorable conditions. For example, polymer can increase the viscosity of water to make it closer to oil viscosity in order to sweep oil out of the reservoir (Zhu et al., 2013). As for surfactant and alkaline, they can lower interfacial tension or IFT between water and oil (Green & Willhite, 1998) that results in increasing either the sweep efficiency or the displacement efficiency. Furthermore, these chemical fluids can be mixed together to increase more efficiency that is called chemical combination flooding or combined flooding such as alkaline-polymer (AP) flooding, surfactant-polymer (SP) flooding, and alkaline-surfactant-polymer (ASP) flooding.

In this study, surfactants are considered as one of the good chemical agents and it is more suitable method to increase emulsion mobility due to lowing oil viscosity and less its acid number. Surfactant can significantly reduce the interfacial tensions between water and oil interfaces (A. Kumar et al., 1984), facilitates emulsion mobility and decreases residual oil saturation. Surfactants or surface active agents are absorbed at a surface or fluid-fluid interface resulting in lower interfacial tension. While the interfacial tension is lower, surface force tends to form one liquid to another single liquid emulsion phase resulting in easily carrying oil up to surface. Interfacial tension are sensitive to pH, pressure, temperature, salinity and surfactant concentration (Green & Willhite, 1998). As the temperature, pressure, and solvent environment of a surfactant (e.g., cosolvent addition, pH changes, or the addition of electrolytes in aqueous systems) vary significant alterations in the solution, various interfacial properties of the surfactant may occur. The modified chemical structure of the surfactant should be needed to maintain a desired degree of surface activity.

However, there has not been any research focusing on new kinds of surfactants at this reservoir. Also, there are challenges that limit the modeling and prediction of efficient surfactant. The effective IFT reduction on surfactant flooding is controlled by several factors: (1) reservoirs conditions (2) degree of surfactant dilution: and (3) surfactant adsorption onto the rock surface (Zhang et al., 2007). These causes are difficult to know how IFT exactly changes under reservoir conditions because reservoir is not ideal conditions, but this research shows one of useful evidences that affect IFT such as the effect of different types of surfactant, surfactant concentration, salinity, pressure and temperature. Also, the new chemical surfactants applied for reducing the interfacial tension will provide fundamental data for making logical surfactant choices and will be varied in various conditions to observe how each parameter is sensitive to surfactants.

Furthermore, the new chemical surfactants which are conducted in this study can be easily found in the chemical industry as waste and the chemical agents market as following:

1. Amines or Ethanolamine (EA) is produced from Ethylene oxide (EO) and primarily comes in the forms of Monoethanolamine (MEA) and Diethanolamine (DEA). Ethanolamine acts as a weak base and is also used as an ingredient in many used daily production such as shampoos, conditioners, detergents, pharmaceutical products, corrosion inhibitors, fabric softeners, and cosmetics. (Dow, 2003)

2. Fatty alcohol and Glycerin are downstream products of crude palm oil. Fatty alcohol can be used as feedstock in the production of shampoo, dishwashing detergent, cleaning solutions, as well as chemical in the textile industry. Fatty alcohols are mainly used in the production of detergents and surfactants. Glycerin or Glycerol is used in medical, pharmaceutical and personal care preparations, mainly as a means of improving smoothness, providing lubrication and as a humectant. (Salager, 2002)

1.4 Objectives

This research is conducted with the following objectives:

1. To measure the interfacial tension of oil and brine by using rising drop method
2. To screen the effective surfactants used for chemical oil recovery

3. To investigate the effect of pressure, temperature, salinity and surfactant concentration on interfacial tension measurement

1.5 Expected benefits

1. This study will provide the interfacial tension data as fundamental data which can be applied to real operational conditions.

2. The results can be used for future studies such as chemical flooding in core sample and solvent selection for the surfactant flooding test to increase oil recovery efficiency.

In the following sections, the theory and literature review used in this study are described in Chapter 2. Chapter 3 will explain how to do the experiment and what conditions are used. In Chapter 4, the results from IFT measurement will be presented and also discussed about the effect of various parameters on IFT such as pressure, temperature, surfactant concentration, salinity, and divalent ions. The conclusions of this study will be provided in the Chapter 5, and in this chapter also provide the recommendation for future study.

CHAPTER 2

THEORY AND LITERATURE REVIEW

This chapter will explain the fundamental theory of the interfacial tension, characteristic of surfactant and literature reviews are also presented.

2.1 Theory

2.1.1. Interfacial tension

Interfacial tension can be described in term of two immiscible liquids in contact with each other as shown in Figure 2.1

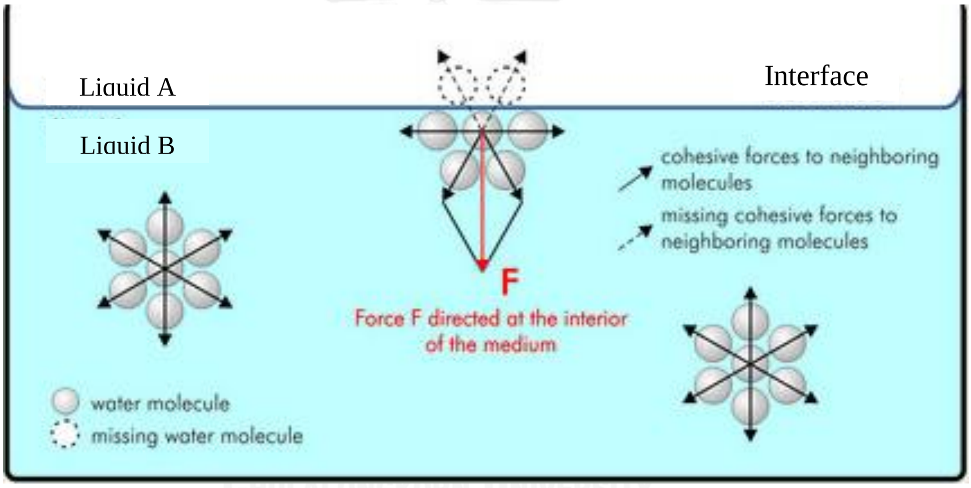

Figure 2.1 Interface between two fluids (GmbH, 2017).

The molecules at the surface between these liquids experience unbalanced forces of attraction. As a result, a force (F) develops that is directed at the interior. These unbalanced forces at the surface of two immiscible fluids (i.e., at the interface) give rise to interfacial tension. (Ghosh, 2009).

Interfacial tension can be defined in the same way as the surface tension. Surface tension is the one of physical property of the liquid surface that allows it to resist an external force, due to the cohesive nature of the liquid molecules.

Antonoff's rule (Antonoff, 1942) predicts that the interfacial tension (γ_{AB}) between two liquids A and B is equal to the difference between the respective surface tensions (i.e., γ_A and γ_B).

$$\gamma_{AB} = |\gamma_A - \gamma_B| \tag{2.1}$$

Therefore, it can be anticipated from this rule that the interfacial tension should lie between the surface tensions of the two liquids.

Young Laplace writes equation to give an expression for the different pressure over an interface between two fluids in terms of the surface tension σ and the principal radii of curvature, R1 and R2. (Svein, 2012)

$$\Delta P = \gamma\left(\frac{1}{R_1} + \frac{1}{R_2}\right) \tag{2.2}$$

$$\Delta P = \frac{2\gamma}{R} \qquad \text{For Spheres} \tag{2.3}$$

$$\Delta P = \frac{\gamma}{R} \qquad \text{For Cylinders} \tag{2.4}$$

The expression is a geometry factor. At equilibrium, each point on the interface has the same geometry factor. The simple expression reflects the fact that for an arbitrary, smooth surface, the curvature at any point is defined by assigning radii of curvature, R1 and R2, in two planes, called principal curvature sections.

The significance of interfacial tension (IFT) is that if the IFT is reduced by chemicals surfactants, oil and water as two phases liquid can be formed to a single liquid. Thus, emulsion which is trapped with the rock can be easily mobilized after flooding with water.

2.1.2 Surfactant

The properties and applications of surfactants are determined by the balance between the hydrophilic and hydrophobic portions of the molecules. The desired properties will vary significantly for applications noted. It seems obvious that our need to understand the relationship between the chemical structures of those materials and their physical manifestations in particular circumstances becomes more important.

Surfactant can be classified into four basic groups depending on the nature of polar head group as anionic, cationic, non-ionic, and amphoteric.

Anionic: This surfactant is called as anionic because it has negative charge on its head group. These are widely used in chemical EOR processes because they exhibit relatively low adsorption on sandstone rock that has negative charge.

Nonionic: This group does not ionize on its head group and the tail group is smaller than a head. Although they are more enduring of high salinity, their function is not as good as anionic to reduce IFT. Nonionic surfactants are used as dispersants. Water hardness has no effect on their function (Zhao et al., 2004).

Cationic: Their head group of this surfactant has a positive charge. Cationic are able to strongly adsorb in the sandstone rocks.

Zwitterionic: When any surfactants present both positive and negative charge is called zwitterionic.

In aqueous systems, the head group will be ionic or highly polar, so that it can act as a solubilizing functionality. In a nonpolar solvent such as hexane the same groups will, in theory, function in the opposite sense. The unusual properties of aqueous surfactant solutions can be ascribed to the presence of a hydrophilic head group and a hydrophobic chain (or tail) in the molecule. The polar or ionic head group usually interacts strongly with an aqueous environment, in which case it is solvated *via* dipole–dipole or ion–dipole interactions. In fact, Table 2.1 is the nature of the polar head group which is used to divide surfactants into different categories.

Table 2.1 Surfactant classification (Azarmi & Ashjaran, 2015)

Types of surfactant	Example
Anionic	Sulfonation and sulfation
	Alkyl sulfate (Alkyl-Ester-Sulfate)
	Phosphate ester
	Carboxylates
Cationic	Quaternary alkyl-ammoniums
	Fatty amine
Nonionic	Alcohol
	Amine oxide
	Ethoxylated amines
Amphoteric	Carboxybetaines
	Sulfobetaines

2.1.3 Surfactants used in this study

In this study, nonionic surfactants are conducted to measure the interfacial tension between oil and water. Although their function is not as good as anionic to reduce IFT, they are more enduring of high salinity in Northern oilfield conditions. All nonionic surfactants are the waste products from carbon dioxide capture process and from chemical company. Therefore, if all surfactants can be used for increasing emulsion mobility effectively, all surfactants will have more valuable. Also, all of them are quite cheap and easy to find in chemical market. These are reasons why all surfactants are chosen in this study.

The term nonionic surfactant usually refers to derivatives of ethylene oxide with an alcohol containing an active hydrogen atom (Hepworth, 2006). However, other types such as alkanolamines, amine oxides, fatty acids, and fatty amines are all produced and used widely throughout the world.

1.) Ethanolamine group

Ethanolamine is an amino alcohol. Ethanolamine is produced commercially by animating ethylene oxide with ammonia; the replacement of hydrogen of ammonia with an ethanol group produces ethanolamine.

Ethanolamine is reactive and bifunctional, combining the properties of alcohols and amines. Ethanolamine is produced by reacting 1 mole of ethylene oxide with 1 mole of ammonia. Typically, ethylene oxide is reacted with ammonia in a batch process to produce a crude mixture of approximately one-third each ethanolamine, diethanolamine, and triethanolamine, which is then separated, achieving varying degrees of single component purity. Figure 2.3 shows why both monoethanolamine and diethanolamine are the base form, and Figure 2.4 shows pH value of ethanolamine solution.

- Monoethanolamine (MEA) C_2H_7NO

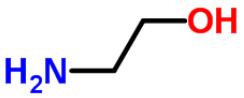

Figure 2.2 Chemical structure of MEA (Wilma & Bergfeld, 2012)

- Diethanolamine (DEA) $C_4H_{11}NO_2$

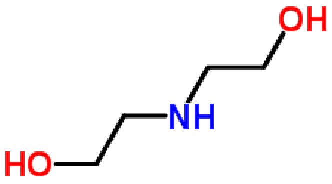

Figure 2.3 Chemical structure of DEA (Wilma & Bergfeld, 2012)

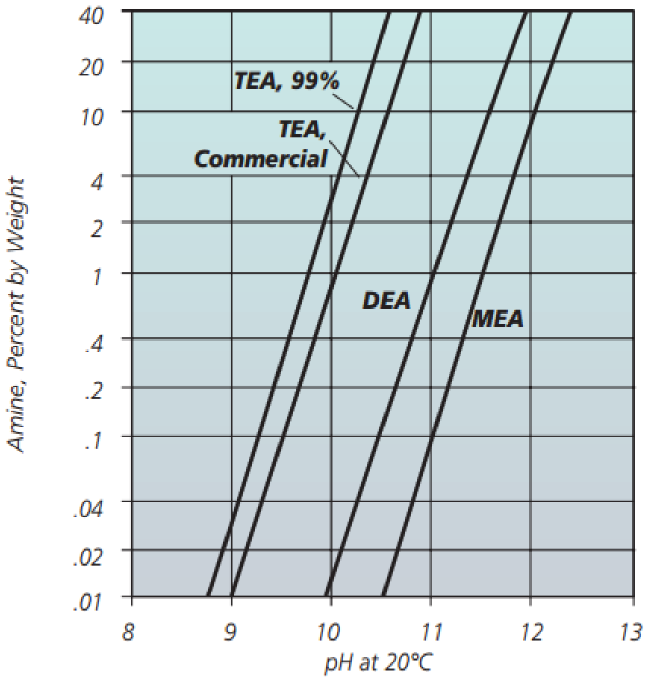

Figure 2.4 pH of ethanolamine solution (Dow, 2003)

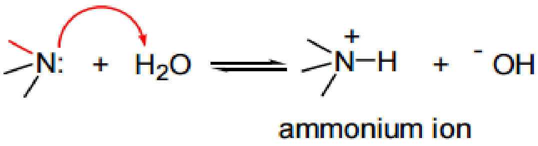

Figure 2.5 Chemical reaction between ethanolamine and water (Tangpasutadon, 2010)

2.) Fatty alcohol and glycerine group

The hydroxyl group is a functional group that consists of a hydrogen atom covalently bonded to an oxygen atom. The hydroxyl group is denoted by -OH in chemical structures and has a valence charge of -1. When the hydroxyl group is the dominant functional group in an organic compound, that compound behaves as an alcohol.

The electronegativity of oxygen is substantially greater than that of carbon and hydrogen. Therefore, the covalent bonds of this functional group are polarized in order that oxygen is electron rich and both carbon and hydrogen are electrophilic.

- Fatty Alcohol or FAOH C0898 (Octyl Alcohol) $CH_3(CH_2)_7OH$.

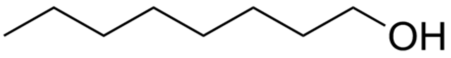

Figure 2.6 Chemical structure of Fatty Alcohol C0898 (Schonfeldt, 1969)

- Fatty Alcohol or FAOH C1098 (Decyl Alcohol) $CH_3(CH_2)_9OH$

Figure 2.7 Chemical structure of Fatty Alcohol C1098 (Schonfeldt, 1969)

- Fatty Alcohol or FAOH C0810 (Octyl Decyl Alcohol) $C_{18}H_{38}O$

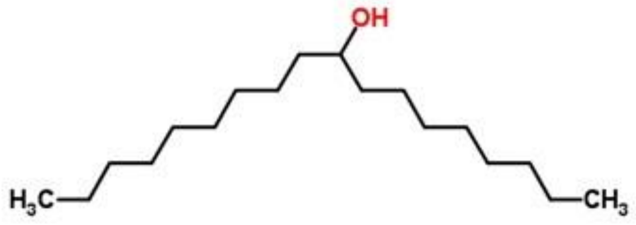

Figure 2.8 Chemical structure of Fatty Alcohol C0810 (Schonfeldt, 1969)

- Fatty Alcohol or FAOH C1214 (Lauryl-Myristyl Alcohol)

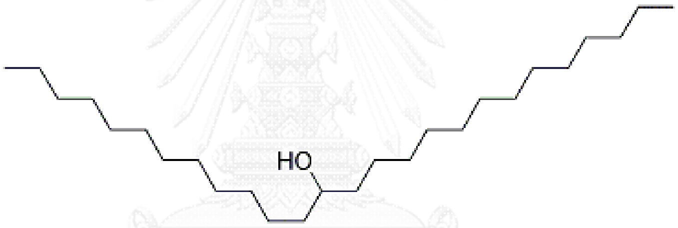

Figure 2.9 Chemical structure of Fatty Alcohol C1214 (Schonfeldt, 1969)

- Refined glycerine $C_3H_8O_3$

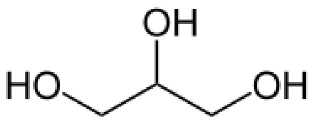

Figure 2.10 Chemical structure of refined glycerine (Schonfeldt, 1969)

2.1.4 Phase behavior

The mechanism for IFT reduction in aqueous solution can be described in term of colloidal aggregates of surfactant molecules which are called micelles. When the surfactant is put in aqueous solution, the dissolved surfactant molecules are dispersed on the interface as monomers and the surface free energy has been decreased. After that, when the surfactant concentration is added more until the specific surfactant concentration called critical micelle concentration (CMC) is reached, the surfactant start aggregating into micelles as shown in Figure 2.9 (Sheng, 2011), and surface tension are plotted as a function of concentration in Figure 2.10 (Attwood & Florence, 2012). At above critical micelle concentration, the additional surfactants will be insignificant on reduction of interfacial tension and also excess amount of surfactant in aqueous solution.

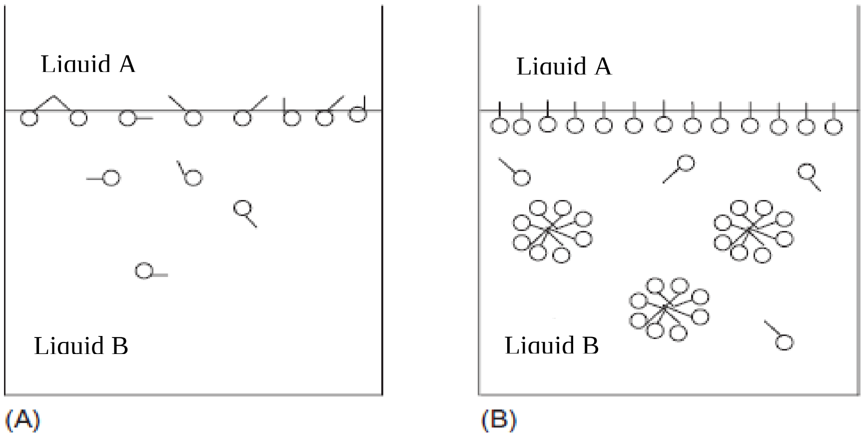

Figure 2.11 Distribution of surfactant molecules in solution (A) below CMC and (B) above CMC (Sheng, 2011)

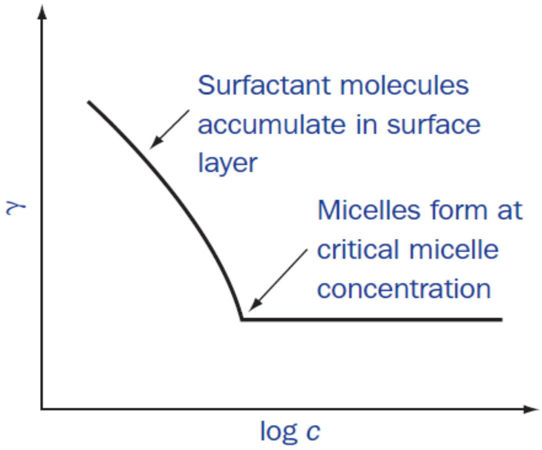

Figure 2.12 Relationship between surface tension and surfactant concentration (Attwood & Florence, 2012)

The CMC values are used in actually process industry surfactant applications such as mineral processing to determine of personal care food, to separate the different type of plastic and to bring new surfactant into proper form because the greatest surfactant effect will be accomplished when significant micelles concentration is propose (Schramm et al., 2003). Thus, the CMC is very important to concern when there are discussion in the formulation of micelles and applications in many process surfactant industries.

The length of the chain of a hydrocarbon surfactant has been a factor effecting on CMC. It is known that the CMC decreases logarithmically as the number of carbons in the chain of a homologous series increases (Myers, 2006). For straight chain hydrocarbon, the CMC is usually reduced with the addition of each –CH2– group. This is called Traube’s rule (Attwood & Florence, 2012). The increase of hydrophobic tail length results in surfactants that are more efficient. Also, the presence of polar substituent groups on the hydrophobic chain can produce different effects on the CMC.

2.2 Literature reviews

Bera et al. (2011) studied the interfacial tension and the phase behavior of surfactant-brine–oil system that was concerned in the industry to screen surfactants and their formulations for reducing IFT in aqueous phase behavior. In this study, the oil-water phase containing different ethoxylated (nonionic commercial grade surfactants), secondary alcoholic surfactants and brine (NaCl) were contacted with synthetic oil to observe their phase behavior. Results showed that the interfacial tension between oil and microemulsion phase was a strong function of both concentration of surfactant and salinity. The results of micelle concentration (CMC) of different three surfactant (15-S-5, 15-S-7 and 15-S-9) solutions were 0.2, 0.3 and 0.4 concentration of surfactant wt.% x 10^{-2} respectively. They confirmed that after CMC value the concentration of surfactant became more stable. Another result was testing to observe relationship between salinity and interfacial tension. As salinity was added, the interfacial tension of excess oil microemulsion decreased, while excess brine microemulsion system increased.

Ahmadi et al. (2014) studied the new surfactant to reduce interfacial tension between oil and water. They extracted a natural based surfactant from the leaves of the mulberry tree. The results showed that when a natural based surfactant increased 1 wt.% concentration of micro-sized mulberry leaf particles, it could decrease the IFT of a system consisting of distilled water and kerosene by 60% from 44 dyne/cm to 17.9 dyne/cm.

The phase behavior and interfacial tension of new designed surfactant on heavy oil, and the effect of salinity were investigated (Dehghan et al., 2015). In this research, surfactants concentration varied at 0.05, 0.1, 0.2, and 1 wt.%, salinity ranging 20,000-250,000 ppm were measured. As the results indicate that the interfacial tensions was declined with increasing the surfactant concentration in all cases. In all cases of the surfactant concentrations were less than 1 mN/m at salinities more than 40,000 ppm. The interesting phenomena in these series of the experiments was the existence of the lowest IFT value, less than 0.07 mN/m, at salinities around 150,000 ppm. Thus, it could prove from the hypothesis that middle phase micro-emulsion had lowest IFT. One more interesting was the small raising interfacial tension values with increasing the salinities to the water in oil emulsion. The main reason was a raise in the repulsive electrostatic forces between the phase molecules. This phenomenon deteriorated the mass transfer between the phases, resulting in the interfacial tension began to increase.

Karnanda et al. (2013) studied the various parameters that effect on the interfacial tension such as temperature, pressure, salinity, and surfactant concentration. The pressure was varied from atmospheric to 4,000 psi. Several conditions in aqueous phases were prepared, i.e., distilled water, 10% brine (100% NaCl), 10% brine (95% NaCl and 5% $CaCl_2$), and 10% (83% NaCl and 17% $CaCl_2$). There were only three surfactants of 13 commercial surfactants showed good solubility in pure water and brine. These were called Zonyl FSE Fluorosurfactant®, Triton X-100®, and Triton X-405®. The results showed effect of each parameter as following:

1. The incremental of surfactant concentration could exponentially decrease IFT. Nonionic surfactants were more effective in reducing IFT compared with the anionic surfactant. At the same concentration

2. Temperature has minor effect on IFT measurements with brine solution, purified water, and anionic Zonyl FSE. More efficient temperature effect was seen for nonionic surfactants solutions with IFT increasing with temperature for Triton X-100 and decreasing for Triton X-405 solutions

3. Pressure had no effect on IFT values except for pure brine where minimal increase on IFT was seen.

4. The presence of salt in the solution can decrease the IFT value of surfactants. The drop is more drastic for Triton X-405 than that for Zonyl FSE.

5. Composition of salt had small effect, especially at low surfactant concentrations. Solutions of salt composition of 95% NaCl and 5% $CaCl_2$ presented the highest drop.

6. Surfactant flood conducted at residual oil saturation was able to produce 4% OOIP more oil than that obtained with conventional water flood.

The effects of ethoxylated nonyl phenols as nonionic surfactants with ethoxy group numbers 4 and 9 (nonyl phenol 4 and nonyl phenol 9) on the interfacial tension between crude oil and water were investigated (Mosayeb & Abedini, 2012). The crude oil applied in this study was obtained from the Refinery of Tehran. The interfacial tension measurements were carried out with rising drop method. Drop formation is performed under controlled temperature and pressure, with the maximum drop size recorded photographically. The results showed that when concentration increases ranges 0 to 1.25 g/l, interfacial tensions of nonyl phenol 4 and nonyl phenol 9 decreased of 27.86 $mN.m^{-1}$ to 1.85 mN/m and 2.63 $mN.m^{-1}$, at 25°C, respectively. For mixture of two surfactants, because of formation of mixed micellar aggregates, mixed surfactants had more influence on reduction of interfacial tension than individual surfactant.

There are many researches working on the type of surfactant-brine-oil system. However, there has not been any research focusing on surfactant flooding as tertiary recovery in Northern oilfield, Thailand. Therefore, the main objective of this study is to find the effective surfactants used for surfactant flooding and to investigate the effects of various parameters on IFT.

CHAPTER 3

METHODOLOGY

In order to measure the interfacial tension measurement, fluid property experiment need to be tested. Therefore, in this study, there are two experiments that would be performed.

3.1 Fluid properties

Oil sample and produced water are received from the Northern oilfield in Thailand. Before both of them are used to determine interfacial tension, they have to be tested to find their composition. For produced water, it is used as the aqueous solution, but each type of surfactant is used as chemical substance.

3.1.1 Oil Composition

Oil composition at Northern oilfield analyzed by Intertek Testing Services (Thailand) Ltd. Oil composition is analyzed by using gas chromatography method. This method examines the complex Alkane distribution of crude oil sample. The oil composition has several kinds of hydrocarbon from C6 to C35+ as shown in Table 3.1

Table 3.1 Oil sample composition from gas chromatography test

Composition	Percent by weight
C7	2.99
C8	7.24
C9	3.49
C10	2.72
C11	2.42
C12	2.32
C13	3.6
C14	4.12
C15	4.34
C16	3.78
C17	4.13
C19H40	1.00
C18	3.16
C20H42	0.37
C19	3.52
C20	3.69
C21	3.76
C22	3.43
C23	3.80
C24	3.45
C25	3.72
C26	3.64
C27	3.84
C28	3.05
C29	3.04
C30	2.60
C31	2.24
C32	1.56
C33	1.22
C34	1.16
C35+	3.51

3.1.2 Oil Density

The oil density is the one of physical properties that is required. The oil sample is heat up to desired temperatures at 70 80 and 90 °C. After that the oil sample is fed into syringe at certain volume and then measured its weight by precision scale meter. The oil density can be computed by dividing mass by volume with equation 3.1 and presented in Table 3.2.

$$\rho = \frac{m}{V} \tag{3.1}$$

Where ρ is density,

m is mass, and

V is volume.

Table 3.2 Oil density at different temperature

Temperature (^{o}C)	Oil density (g/cm^3)
70	0.85165
80	0.84616
90	0.84335

Also, this oil sample is also tested with the acid number of 0.08 mg KOH/g

3.1.3 Brine Properties

Produced water coming out from the reservoir to the surface is analyzed to observe the brine composition as presented in Table 3.3

Table 3.3 Produced water composition from laboratory test

Chemical Ion	Concentration (ppm)
Sodium, Na	6,325
Calcium, Ca	183
Magnesium, Mg	27.7
Potassium	54.3
Chloride, Cl	9,422
Sulfate, SO_4	529
Carbonate, CO_3	Less than 5.0
Bicarbonate, HCO_3	97
Hydroxide, OH	Less than 5.0

As shown in the table, there are two main chemical compositions which are sodium (38.01 %), and chloride (56.63 %). There is a small amount of divalent ion such as calcium and magnesium compared with the main chemical compositions. Therefore, the composition of divalent ion would be ignored for the main experiment. However, the effect of divalent ion is also studied.

3.2 Interfacial tension Measurement

3.2.1 Apparatus

Figure 3.1 shows the interfacial tension meter (Model 700, Vinci Technology) used to measure the interfacial tension (IFT) between liquid-liquid and liquid-gas interface. This equipment has cell volume of 25 cm^3. Also, it can measure the IFT in a range of 0.01 to 72 mN/m and its accuracy is within 0.01 mN/m. The maximum working pressure and temperature are 69 MPa (or 10,000 psi) and 180°C (or 350°F) respectively. A calibrated capillary into a bulk fluid in a cell created an oil drop. Then, a computer connected with camera would analyze the IFT value and also recorded the shape of the drop with Drop Analysis Software (DAS) provided by Vinci Technology.

Figure 3.1 IFT 700

3.2.2 Procedure

Firstly, the simulated brine was prepared from sodium chloride and distilled water at desired salinity and surfactant concentration. The IFT 700 was set up to run the experiment. The running process such as chamber, needle, pipe system, and pump would be cleaned by using acetone and distilled water. Next, all the components of those would be combined and checked the vertical setup.

Before measuring the IFT, software required both density of oil and surfactant solution. The density of surfactant solution could be measured by density meter. Also, the camera had to be adjusted to create the bright resolution.

After that, both oil sample and surfactant solution were separately fed into the cylinder so as to start this experiment. Both of them were heated up to the desired temperature which could be set up at the software. To reach equilibrium the temperature was kept about 20 minutes. Then, the surfactant solution as well as the oil would be injected to the chamber of IFT 700 at designed pressure. The camera connected to computer would detect the generated drop which was created by calibrated capillary in the chamber as presented in Fig 4.2. The results would then be recorded by Drop Analysis Software (DAS).

Figure 3.2 Rising oil drop

Before investigating the effect of each parameter on IFT, The screening process is required to search for the best 4 out of 7 surfactant samples that are presented below:

- Monoethanolamine (MEA)
- Diethanolamine (DEA)
- Fatty Alcohol (Octyl Alcohol) or FAOH C0898
- Fatty Alcohol (Octyl-Decyl Alcohol) or FAOH C0810
- Fatty Alcohol (Lauryl-Myristyl Alcohol) or FAOH C1214
- Fatty Alcohol (Decyl Alcohol) or FAOH C1098
- Refined Glycerine (RG)

All of these types of surfactant are tested at the base case reservoir conditions: pressure at 1,500 psi, temperature at 80°C, salinity at 15,000 ppm, and surfactant concentrations at 0.05 %wt. After the results are compared with each other, the best 4 out of 7 surfactant samples will be decided and then four samples will be investigated in the future.

There are 4 parameters that are concerned in this study: temperature, pressure, surfactant concentrations, and salinity. Firstly, there are the best 4 kinds of surfactant from screening process which would be prepared at different designed concentrations. Secondly, the salinity varied from 7,500, 10,000, 15,000 to 17,500 ppm would be investigated with surfactant solution at various concentrations. Thirdly, the temperature is also ranged at 70°C, 80 °C, and 90°C. Lastly, pressure would be pumped for 3 values at 1,000, 1,500, 2,000 psi. The operating conditions for this study are presented in Table 3.4. Also, the flow chart of the study is illustrated in Figure 3.3.

Table 3.4 The operating conditions in this experiment

Surfactants	Surfactant Concentration (%wt.)	Salinity (ppm)	Temperature (°C)	Pressure (psi)
-Monoethanolamine - Diethanolamine - Refined Glycerine	0.00	0	70	1,000
	0.50	7,500	80	1,500
	1.00	10,000	90	2,000
	2.00	15,000		
	4.00	17,500		
Fatty Alcohol C1098	0.00	0	70	1,000
	0.25	7,500	80	1,500
	0.50	10,000	90	2,000
	0.75	15,000		
		17,500		

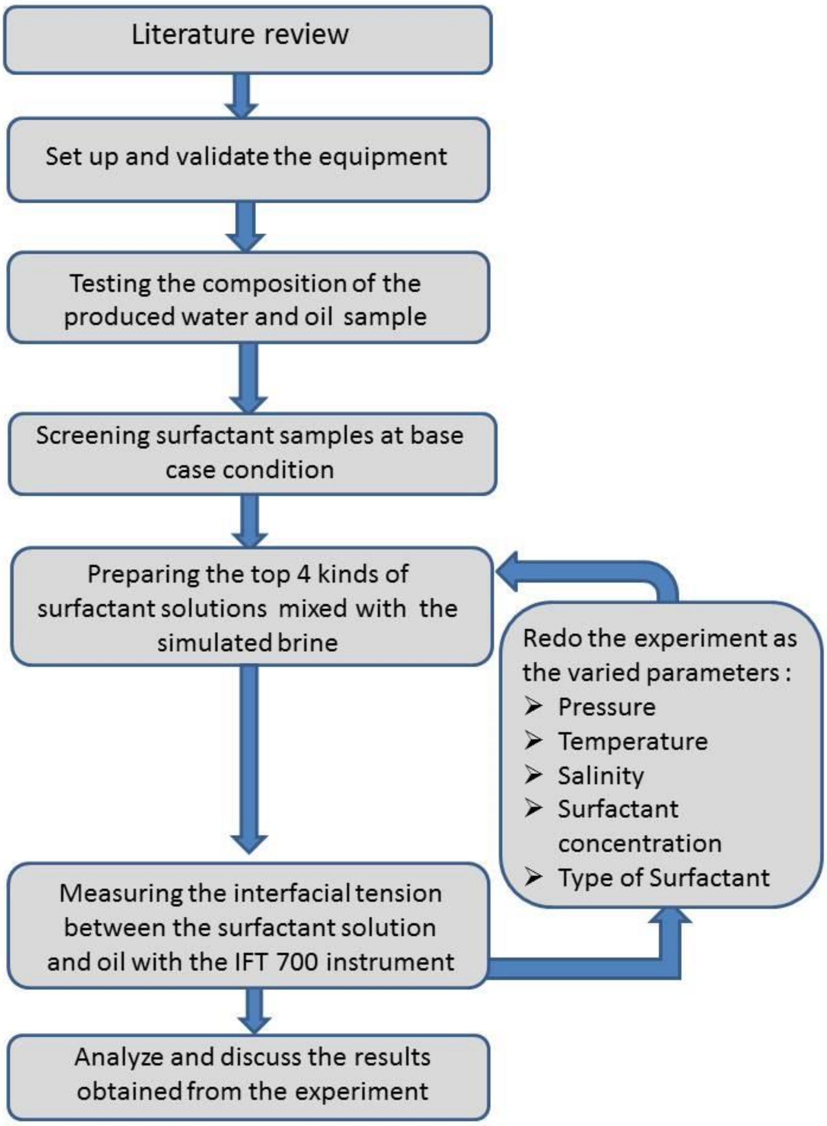

Figure 3.3 Methodology flow chart

CHAPTER 4

RESULTS AND DISCUSSION

The results of the effects of various parameters such as pressure, temperature, concentration of surfactants, type of surfactants, salinity and divalent ions on interfacial tension of surfactant solution are presented and discussed in this chapter.

4.1 Verification of equipment and procedure

The equipment used in this experiment is verified by comparing the result from the previous work (Saengnil, 2015) and (Asavaritikrai, 2016) before running the experiment of this study. Saengnil and Asavaritikrai conditions are at 0.05 wt.% of sodium hydroxide, temperature of 80 °C and salinity of 750 ppm. Pressure was varied at 500, 1,000 and 1,500 psi. The result of equipment verification was present in Figure 4.1 and Table 4.1

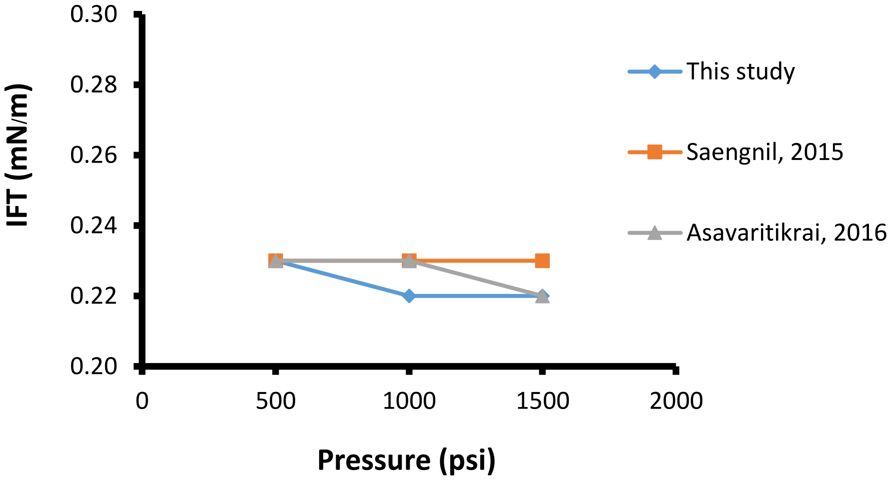

Figure 4.1 Verification of equipment and procedure (NaOH concentration = 0.05 wt.%, temperature = 80 °C, salinity = 750 ppm)

Table 4.1 Verification of equipment and procedure (NaOH concentration = 0.05 wt.%, temperature = 80 °C, salinity = 750 ppm)

IFT (mN/m)	Pressure (psi)		
	500	1,000	1,500
Saengnil (2015)	0.23	0.23	0.23
Asavaritikrai (2016)	0.23	0.23	0.22
This study	0.23	0.22	0.22
% Error	0	4.3	4.3

The result of the verification shows that the IFT value from the current study was similar to the IFT value from the Saengnil (2015) and Asavaritikrai (2016) at the same conditions, with the maximum error of 4.3%. As for percent error, the maximum error was less than 5%; thus, the equipment of this study was valid to be employed in this research.

4.2 Screening process

The screening process is required to find the best 4 out of 7 surfactant samples: monoethanolamine (MEA), diethanolamine (DEA), fatty Alcohol (Octyl Alcohol) or FAOH C0898, fatty Alcohol (Octyl-Decyl Alcohol) or FAOH C0810 fatty Alcohol (Lauryl-Myristyl Alcohol) or FAOH C1214, fatty Alcohol (Decyl Alcohol) or FAOH C1098, and refined Glycerine (RG). The best 4 of all surfactants have to have a small IFT value than others.

All of these types of surfactant are tested at the base case reservoir conditions: pressure at 1,500 psi, temperature at 80°C, salinity at 15,000 ppm, and surfactant concentrations at 0.05 %wt. As shown in Table 4.2 and Figure 4.2, the results show that the best 4 of all surfactants are monoethanolamine (MEA), diethanolamine (DEA), fatty Alcohol (Decyl Alcohol) or FAOH C1098, and refined Glycerine (RG). Based on the IFT reduction, these 4 surfactants will be selected to further study.

Table 4.2 Screening results at surfactant concentration 0.05 wt.%, salinity 15,000 ppm, pressure 1,500 psi

IFT (mN/m)	70 °C	80 °C	90 °C
MEA	22.95	21.88	19.85
DEA	23.55	22.71	20.35
FAOH C1098	41.10	40.69	39.86
RG	43.81	42.44	40.41
FAOH C0898	43.83	42.88	41.91
FAOH C1214	44.16	42.52	41.69
FAOH C0810	44.25	43.25	42.88

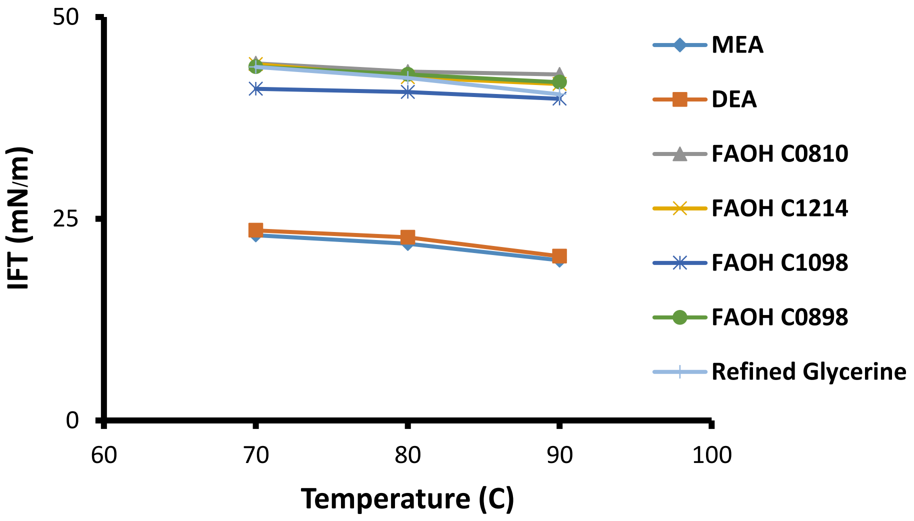

Figure 4.2 Screening process at the base case reservoir conditions

4.3 The effect of pressure on the interfacial tension

The pressure applied in this study covered the pressure used in Northern oilfield in Thailand from 1,000 to 2,000 psi. The results are shown in Figure 4.3, the percentage change of interfacial tension in distilled water, Monoethanolamine (MEA), Diethanolamine (DEA), Fatty alcohol (FAOHC1098) and Refined Glycerine (RG) are approximately 0.14, 0.91, 0.13, 0.27, and 0.19, respectively.

From the results, when the pressure is varied from 1,000 to 2,000 psi, the interfacial tension is relatively stable because this study is focused on the liquid phase system. Basically, the liquid phase system has more intermolecular force than gas phase system. Thus, the pressure has less effect on this system.

In addition, from the previous studies (Green & Willhite, 1998), it is shown that the pressure on phase behavior of liquid at constant temperature has a small effect on IFT reduction. Also, the interfacial tension would have just slightly changed with pressure at constant temperature (Saengnil, 2015). Therefore, the pressure ranging from 1,000 psi to 2,000 psi at constant temperature can be concluded that it had less significant effect on IFT reduction and it could be ignored for other conditions. The test of this study will apply only pressure at 1,500 psi.

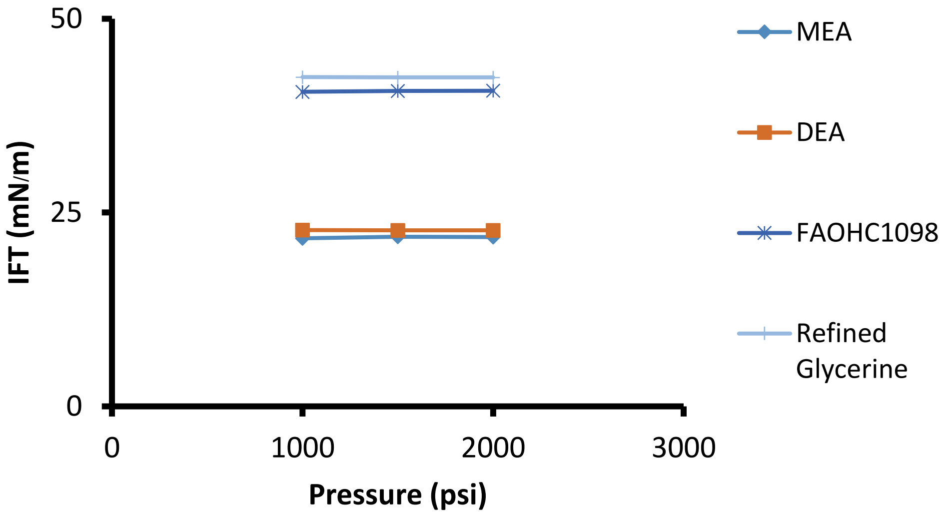

Figure 4.3 Effect of pressure on interfacial tension reduction at 0.05 wt.%, salinity 15,000 ppm, and 80 °C

Table 4.3 Effect of pressure on interfacial tension reduction
at surfactant concentration 0.05 wt.%, salinity 15,000 ppm, and 80 °C

Solutions	IFT (mN/m)		
	1,000 psi	1,500 psi	2,000 psi
Distilled water	43.49	43.55	43.56
Monoethanolamine	21.68	21.88	21.85
Diethanolamine	22.74	22.71	22.64
Fatty alcohol C1098	40.58	40.69	40.71
Refined glycerine	42.52	42.44	42.42

4.4 The effect of temperature on the interfacial tension

The temperature of this study is varied from 70°C to 90°C which is the temperature corresponding to that of the Northern oilfield in order to investigate how temperature impact on IFT, the results of experiment are presented in Figure 4.4 to Figure 4.8 and Table 4.3. From Table 4.3, it is clear that the IFT decreases as the temperature increases.

There are several reasons that can explain the reduction of IFT mechanisms. According to Wei (2005), when the temperature is higher, the free energy between oil and solution will reduce thus resulting in the enhanced mobility of emulsion and also decreasing the IFT. One more reason is that the increment of temperature can affect surfactant solution because of the weakening of intermolecular forces at the oil-water interface. Furthermore, the increasing of temperature led to increase mutual solubility of the solvents, diffusion velocity of surfactant molecules onto the interface, and adsorption velocity of surfactant molecules at the interface, thus reducing the IFT (Mosayeb & Abedini, 2012).

Table 4.4 Effect of temperature on the IFT (surfactant concentration = 0.5 wt.%, , salinity = 15,000 ppm, pressure = 1,500 psi)

IFT (mN/m)	Temperature (°C)		
	70	80	90
Brine without surfactant	44.45	43.55	43.23
MEA	12.73	11.66	11.01
DEA	13.88	13.37	13.04
FAOHC1098	31.53	31.19	30.57
Refined glycerine	43.30	41.96	39.71

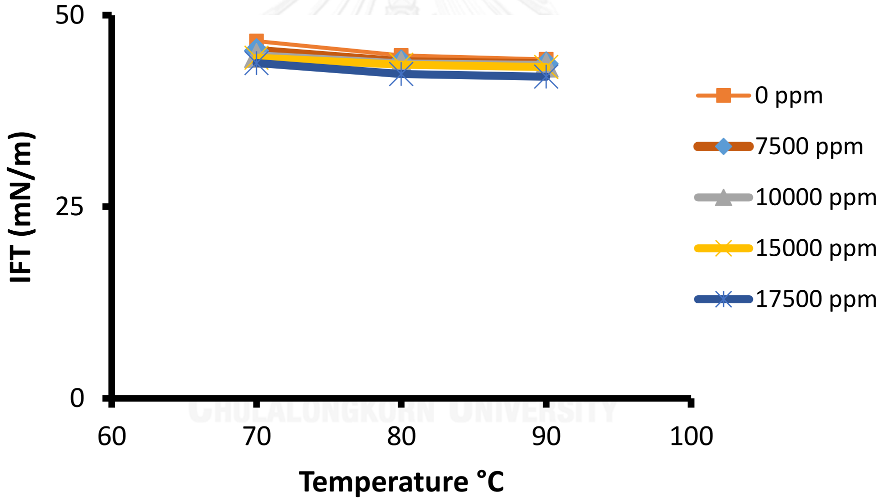

Figure 4.4 Effect of temperature of solution without surfactant (pressure 1,500 psi, salinity at 0 ppm, 7,500 ppm, 10,000 ppm, 15,000 ppm, and 17,500 ppm)

From Figure 4.4, in case of distilled water and brine at different salinity without surfactant, the IFT is relatively constant because there is no surfactant or any chemical that causes in-situ surfactant in the aqueous phase. The main compositions of brine on produced water are sodium ion and chlorine ion that are considered as neutral substance. This explains why the percentage of IFT reduction for solution without brine is quite low about 2.0% to 0.7 %.

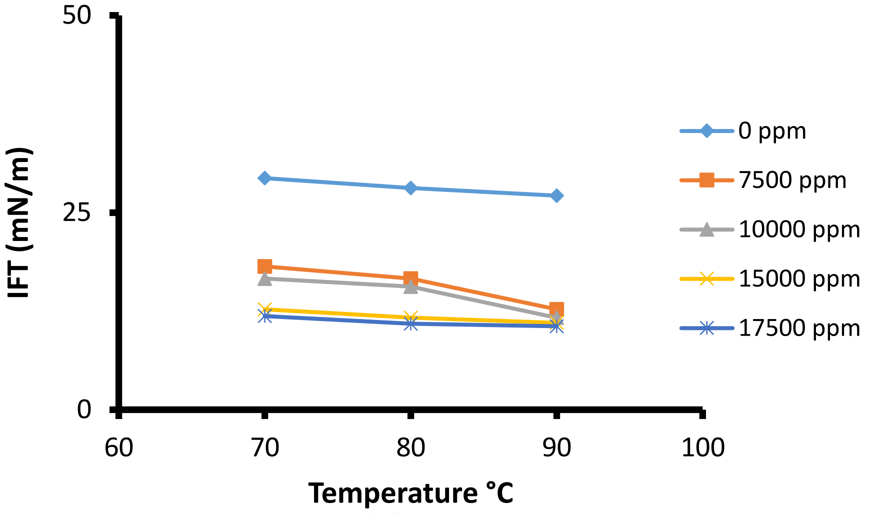

Figure 4.5 Effect of temperature of MEA solution (surfactant concentration = 0.5 wt.%, pressure = 1,500 psi)

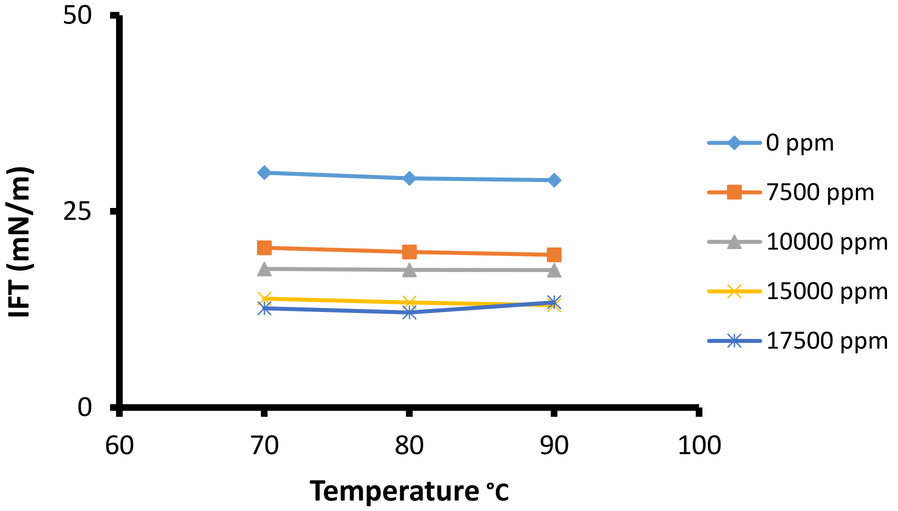

Figure 4.6 Effect of temperature of DEA solution (surfactant concentration = 0.5 wt.%, pressure = 1,500 psi)

However, from Figure 4.5 and 4.6, when the surfactant solution of MEA and DEA which is the base substance is presented in liquid-liquid interface, the increase in temperature can decrease the IFT. The rate of IFT change in case of 15,000 ppm with 0.5%wt. of MEA, temperature ranging from 70°C to 80°C and 80°C to 90°C are 9.18% and 5.9%, respectively whereas the percentage change for DEA solution at the same conditions ranged from 3.81% to 2.53%.

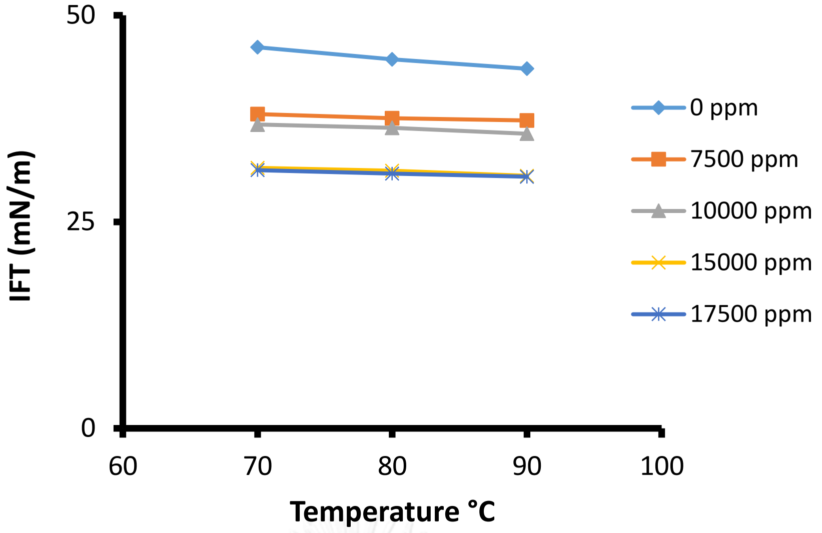

Figure 4.7 Effect of temperature of FAOHC1098 solution (surfactant concentration = 0.5 wt.%, pressure = 1,500 psi)

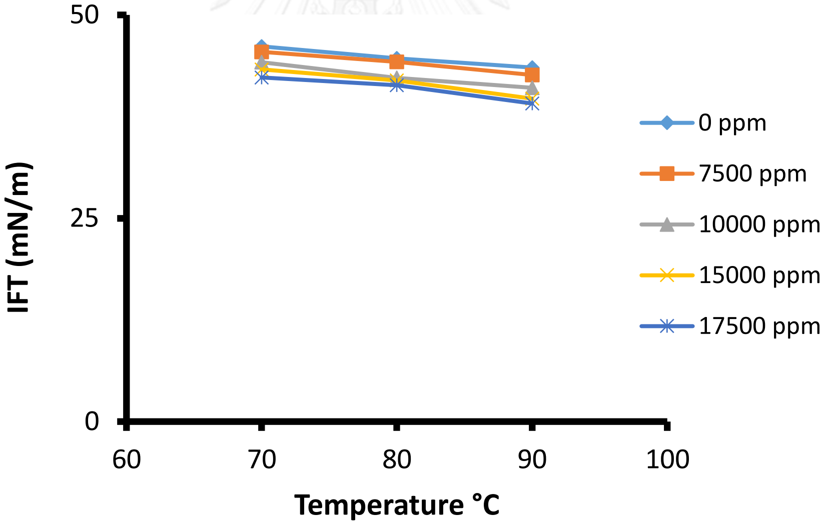

Figure 4.8 Effect of temperature of RG solution (surfactant concentration = 0.5 wt.%, pressure = 1,500 psi)

The effect of temperature in the case of fatty alcohol and refined glycerine can be seen in Figure 4.7 and Figure 4.8, respectively. For Fatty alcohol solution, the percentage of IFT reduction at 70°C to 80°C is 1.1% while the percent different at 80°C to 90°C is 2.03%. For refined glycerine solution, when temperature increases, the percentage of the IFT reduction ranging from 70°C to 80°C and 80°C to 90°C are 3.19% to 5.67%, respectively. The effect of temperature in both solutions gets along well with the results from the surfactant base solution. The increment of temperature may help the fatty alcohol and refined glycerine to dissociate more than at higher temperature.

4.5 The effect of type of surfactant solution on the interfacial tension

After screening the surfactant solution to find the best 4 kind of all surfactant samples, these 4 different types of surfactant which are monoethanolamine (MEA), diethanolamine (DEA), fatty alcohol (C1098), and refined glycerine are used to investigate for the further study of the effects on IFT as shown in Table 4.4 to Table 4.8 and Figure 4.9

Table 4.5 Results of IFT of distilled water (mN/m)

IFT (mN/m)	Salinity (ppm)	Temperature (°C)		
		70	80	90
Distilled water	0	46.61	44.75	44.23
	7,500	45.38	43.91	43.64
	10,000	44.76	43.71	43.51
	15,000	44.45	43.55	43.23
	17,500	43.76	42.34	41.97

Table 4.6 Conc. 0.05 %wt., salinity at 15,000 ppm, pressure 1,500 psi

Surfactant solutions	Temperature (°C)		
	70	80	90
MEA	22.95	21.88	19.85
DEA	23.55	22.71	20.35
FAOH C1098	41.10	40.69	39.86
RG	43.88	42.44	40.41
Without surfactant	44.45	43.55	43.23

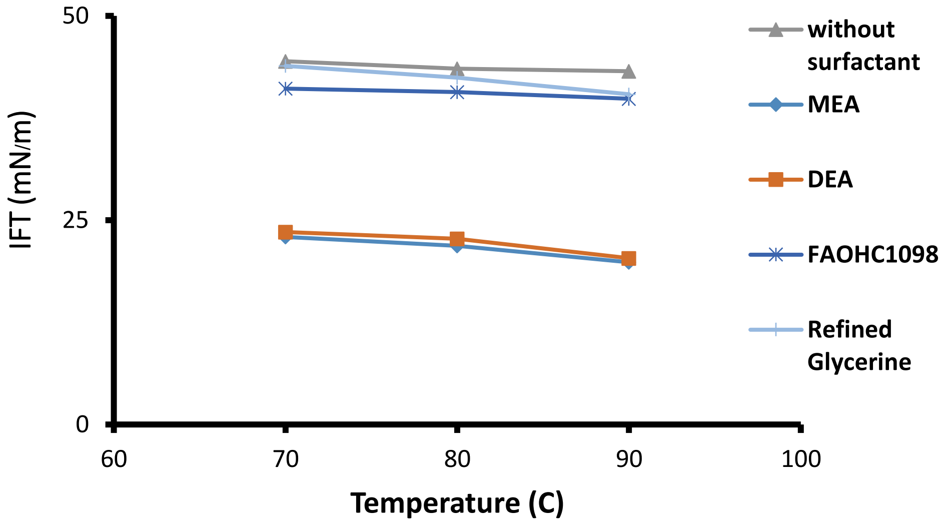

Figure 4.9 The effect of different types of surfactant (surfactant conc. = 0.05 wt.%, Salinity = 15,000 ppm, and pressure = 1,500 psi)

Table 4.7 Results of IFT of monoethanolamine solution (in mN/m)

Conc. of MEA (wt.%)	Salinity (ppm)	Temperature (°C)		
		70°C	80°C	90°C
0.5	0	29.36	28.09	27.14
	7,500	18.17	17.11	15.31
	10,000	16.63	15.59	14.65
	15,000	12.73	11.66	11.01
	17,500	11.87	10.91	10.58
1.0	0	26.69	25.56	24.38
	7,500	16.68	15.96	14.59
	10,000	15.53	14.71	14.06
	15,000	11.72	10.29	9.77
	17,500	10.68	9.54	8.81
2.0	0	23.33	22.24	21.13
	7,500	12.31	11.73	11.36
	10,000	10.05	9.74	9.01
	15,000	6.01	5.77	5.55
	17,500	5.78	5.46	5.24
4.0	0	18.67	18.29	18.02
	7,500	12.05	11.54	11.21
	10,000	9.86	9.4	8.79
	15,000	5.59	5.48	5.03
	17,500	5.37	5.12	4.72

Table 4.8 Results of the IFT of diethanolamine solution (in mN/m)

Conc. of DEA (wt.%)	Salinity (ppm)	Temperature (°C)		
		70°C	80°C	90°C
0.5	0	29.93	29.21	28.98
	7,500	20.36	19.81	19.46
	10,000	17.67	17.55	17.52
	15,000	13.88	13.37	13.04
	17,500	12.64	12.11	13.40
1.0	0	26.05	25.44	24.83
	7,500	18.23	17.64	17.39
	10,000	14.68	14.45	14.33
	15,000	11.90	11.59	11.23
	17,500	11.72	11.40	13.23
2.0	0	24.96	24.67	24.31
	7,500	15.48	15.22	15.06
	10,000	13.34	13.18	13.01
	15,000	11.22	11.21	11.03
	17,500	11.22	11.09	12.70
4.0	0	24.81	24.51	24.24
	7,500	15.30	14.27	14.07
	10,000	13.21	12.72	12.45
	15,000	11.06	10.86	10.72
	17,500	10.72	10.61	12.43

Table 4.9 Results of the IFT of fatty alcohol C1098 solution (in mN/m)

Conc. of FAOH C1098 (wt.%)	Salinity (ppm)	Temperature (°C)		
		70°C	80°C	90°C
0.25	0	41.03	40.45	39.79
	7,500	38.15	38.69	38.24
	10,000	38.23	37.84	37.45
	15,000	38.04	37.63	37.01
	17,500	37.89	37.48	36.96
0.50	0	39.56	38.97	38.36
	7,500	38.04	37.54	37.27
	10,000	36.78	36.38	35.66
	15,000	31.53	31.19	30.57
	17,500	31.25	30.84	30.46
0.75	0	43.85	42.74	41.33
	7,500	41.40	40.93	40.50
	10,000	38.72	38.17	37.68
	15,000	36.15	35.92	35.61
	17,500	36.14	35.78	35.55

Table 4.10 Results of the IFT of refined glycerine solution (in mN/m)

Conc. of RG (wt.%)	Salinity (ppm)	Temperature (°C)		
		70°C	80°C	90°C
0.5	0	46.13	44.66	43.53
	7,500	45.46	44.21	42.62
	10,000	44.18	43.25	41.04
	15,000	43.30	41.96	39.71
	17,500	42.31	41.35	39.11
1.0	0	44.90	44.17	42.83
	7,500	44.14	43.64	41.88
	10,000	43.54	42.57	39.83
	15,000	41.73	41.14	38.63
	17,500	40.87	39.81	38.46
2.0	0	44.35	43.35	40.59
	7,500	43.39	42.64	39.83
	10,000	41.87	41.33	38.94
	15,000	40.93	40.42	38.27
	17,500	40.21	39.05	38.03
4.0	0	44.32	43.32	40.30
	7,500	43.34	42.05	39.41
	10,000	41.76	41.19	38.72
	15,000	40.42	39.94	38.05
	17,500	39.97	38.88	37.97

For all cases, the results show that if there is a certain amount of surfactant in the aqueous phase, the IFT values can be decreased by using the polar group (hydrophilic part) and non-polar group (hydrophobic part). As for head of surfactant, it will connect between two phase liquids. Thus, in case of none of surfactant, it provides the highest IFT values compared with adding small amount of surfactant solution.

From Table 4.6, at temperature 80°C, the IFT for MEA solution could be reduced up to 49.76%, while the decrease of IFT for DEA solution is 47.85%. The results of MEA and DEA can be differentiated from other surfactants because both MEA and DEA have similar group which consisted of the same molecules of amino and hydroxyl group. As amines, they are weak base and reacted with acids to form salts or soaps which can reduce the IFT (Asavaritikrai, 2016). Moreover, MEA is a stronger base than DEA. According to (S. Kumar et al., 1989), the higher pH of solution, the higher amount of the in-situ surfactant produced and thus lowering the amount of oil. Therefore, the IFT reduction of MEA is better than that of DEA when compared at the same conditions as shown in Figure 4.9.

For both fatty alcohol and refined glycerine, at the concentration of 0.05 wt. %, temperature 80°C, the IFT values are higher than that of amine group because the head of surfactant only presented the hydroxyl group and there is no base substance. Therefore, fatty alcohol and refined glycerine can slightly decrease 6.57% and 2.55% respectively. Specifically, for refined glycerine, the reason is that glycerol or glycerine is even completely soluble in water, but it is insoluble in hydrocarbon (oil) due to its three-hydroxyl groups which can be only dissolved in water. Thus, it cannot perform well as a surfactant to lower the IFT.

4.6 The effect of surfactant concentration on the interfacial tension

The surfactant concentrations on the interfacial tension of this study are prepared from MEA, DEA, fatty alcohol, and refined glycerine and the concentrations in each type are varied at 0, 0.5, 1.00, 2.00, and 4.00 wt.% except for case of fatty alcohol studied at 0, 0.25, 0.50, and 0.75 wt.%. For all cases, when the surfactant concentration increases, the interfacial tension becomes lower. It means that the surfactant concentration have the effect on IFT. The results for all type of surfactants are shown in Figure 4.10 to Figure 4.13.

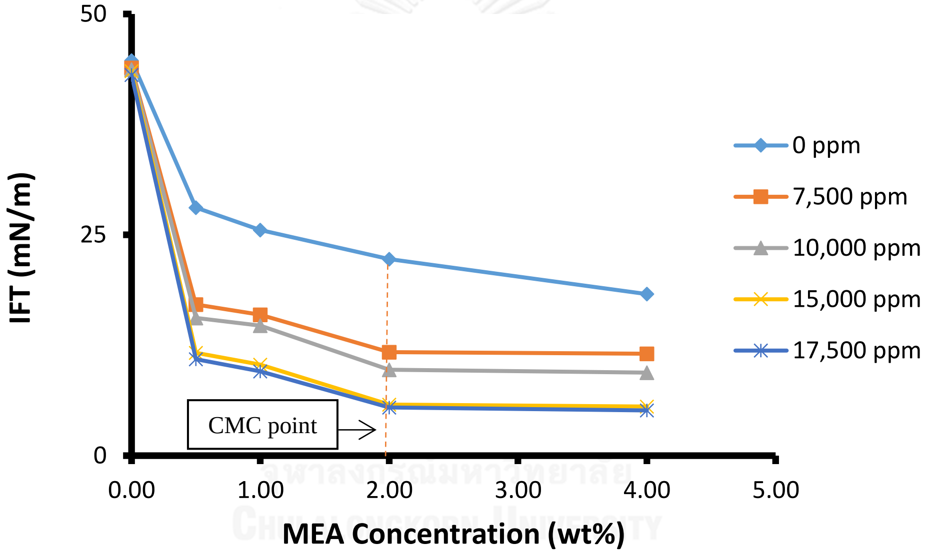

Figure 4.10 Effect of surfactant concentration of MEA solution (tempereature at 80°C, pressure 1,500 psi)

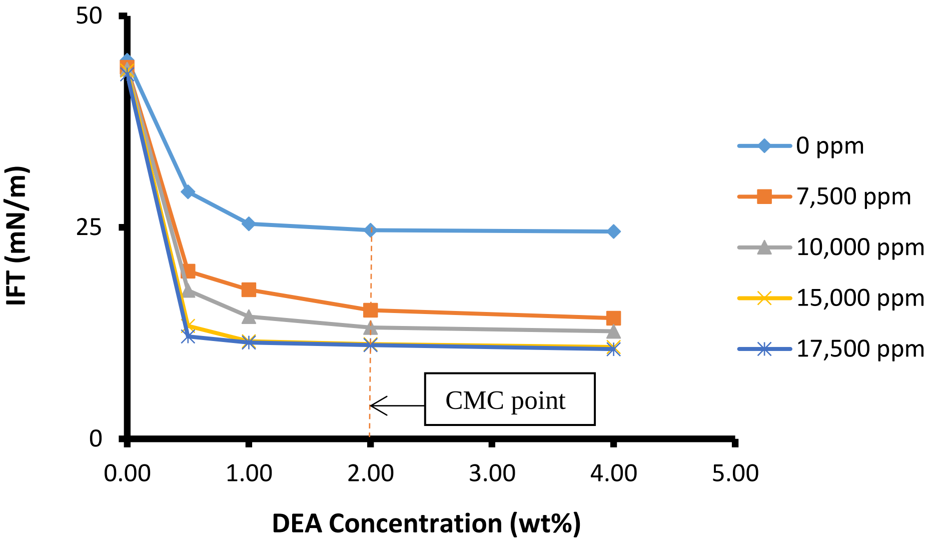

Figure 4.11 Effect of surfactant concentration of DEA solution (tempereature at 80 °C , pressure 1,500 psi)

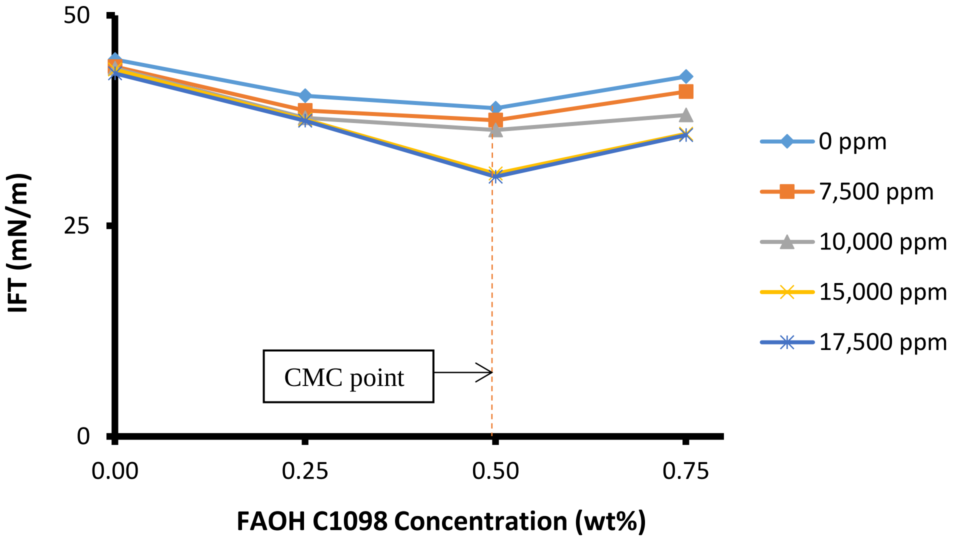

Figure 4.12 Effect of surfactant concentration of FAOH C1098 solution (tempereature at 80 °C , pressure 1,500 psi)

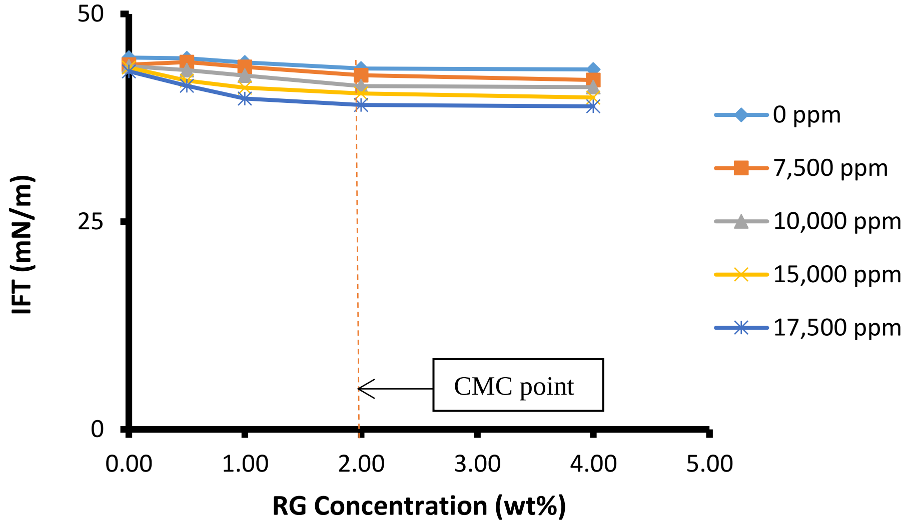

Figure 4.13 Effect of surfactant concentration of RG solution (tempereature at 80 ^{o}C , pressure 1,500 psi)

As shown in Figure 4.10, the effect of MEA concentration is presented at various brine concentrations with pressure of 1,500 psi and temperature of 80°C. The results show that the low concentration varied from 0 to 2.0 wt.% can greatly decrease the IFT up to 87.4%. The reduction of IFT in all cases is caused by molecule of MEA. There are two parts of MEA's molecular structure. One part is called hydrophilic part (hydroxyl and amino groups) which can dissolve in aqueous phase, and the other part is hydrophobic part (hydrocarbon chain) that can dissolve in oil phase. When the surfactant is put into aqueous phase, the interface between oil and water will be covered by the molecules of surfactant as monomers and the surface free energy (surface tension) becomes decreased. Thus, an increase in surfactant concentration can dissolve more both oil and water thus making the reduction of IFT corresponding to (Wu et al., 2014). The surfactant concentration increases until it reaches at 2.0 wt.%, the IFT value may reach its lowest point which is called critical micelle concentration (CMC).

Nevertheless, from Figure 4.10 when the surfactant concentration is higher than 2.0 wt.%, the IFT values become more stable or slightly change in interfacial tension. The reason is that when the surfactant is added in aqueous solution excessively, it can result in the surfactant aggregating into micelles more and more. Consequently, the amount of MEA is difficult to possess in oil-water interface.

For the DEA solution in Figure 4.11, the result show that when the concentration varied from 0 to 2.0 wt.%, the IFT can greatly reduce up to 74.3%. Thus, the increase in surfactant concentration can also decrease the IFT, but it has less efficient to reduce the IFT than MEA. Beyond the CMC, little change in IFT occurs. The surfactant added in excess of the CMC contributes to the formation of micelles and does not increase the concentration at the water/hydrocarbon interface. Therefore, there is only small effect on IFT reduction.

For the fatty alcohol C1098, the mechanism of IFT reduction is the same with MEA and DEA. The results of IFT shown in Figure 4.12 present the relationship between IFT and FAOH concentration. From the Figure 4.12, the IFT value is reduced up to 28.4% until it reaches 0.5 wt.% and becomes slightly increased when FAOH is added in excess of aqueous phase more than 0.5 wt.%. Therefore at 0.5 wt. %, this point can be called CMC point which is a minimum in interfacial tension.

The results of refined glycerine presented in the Figure 4.13 can be described that refined glycerine is ineffective surfactant for the IFT reduction process. Although its molecule has high water solubility, its capability to dissolve in oil is poor. For instance, at salinity 17,500 ppm, 1,500 psi and 80°C, the result showed that the concentrations vary from 0 to 2.0 wt.% can decrease IFT for 9.4%.

Table 4.11 Summary the effect of surfactant concentration

Type of solution	CMC wt.%	% IFT Reduction
Monoethanolamine	2.0	87.4
Diethanolamine	2.0	74.3
Fatty Alcohol C1098	0.5	28.4
Refined Glycerine	2.0	9.4

4.7 The effect of salinity on the interfacial tension

The simulated brine in this study is prepared for composition the same as the produced water from the Northern oilfield with various ions in the reservoir as mention earlier. However, as shown in Table 3.3 the main chemical compositions are sodium ions and chloride ions accounting for 94%. Therefore, the simulated brine used in this study is prepared by mixing sodium chloride and distilled water. The salinity is studied at 7,500, 10,000, 15,000, and 17,500 ppm to investigate the effect of salinity on IFT reduction. The results of the effect of salinity are presented in Figure 4.14 to Figure 4.18.

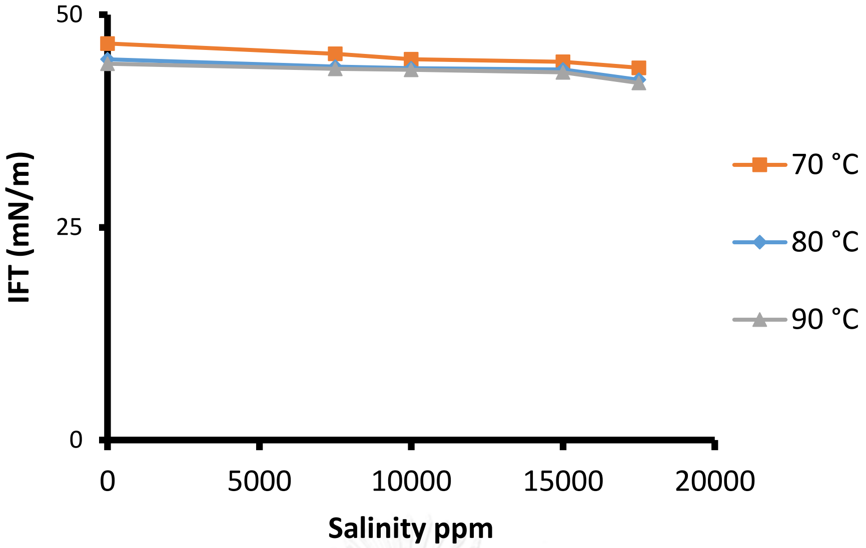

Figure 4.14 Effect of salinity of brine without surfactant substance (pressure 1,500 psi)

From Table 4.5 and Figure 4.14, in cases of brine without surfactant, the interfacial tension is comparatively constant. For example, at temperature 80°C, the IFT can be decreased up to 2.68% when the salinity increased from 0 ppm to 15,000 ppm because there is no amount of surfactant in the aqueous phase. Although the salinity is increased into the aqueous system, there is no amount of surfactant for driving molecule of surfactant to the interface between oil and water. Consequently, the IFT value is slightly decreased at higher salinity.

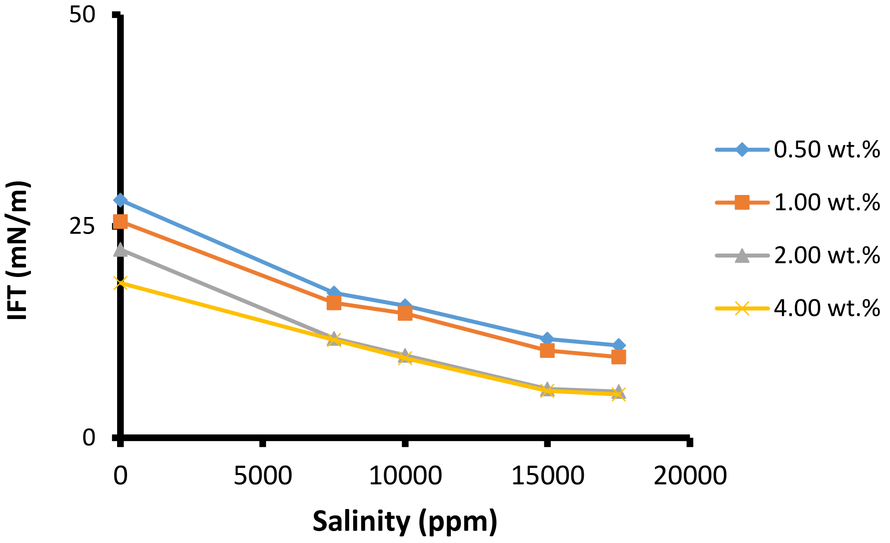

Figure 4.15 Effect of salinity of MEA solution (temperature = 80 °C, pressure = 1,500 psi)

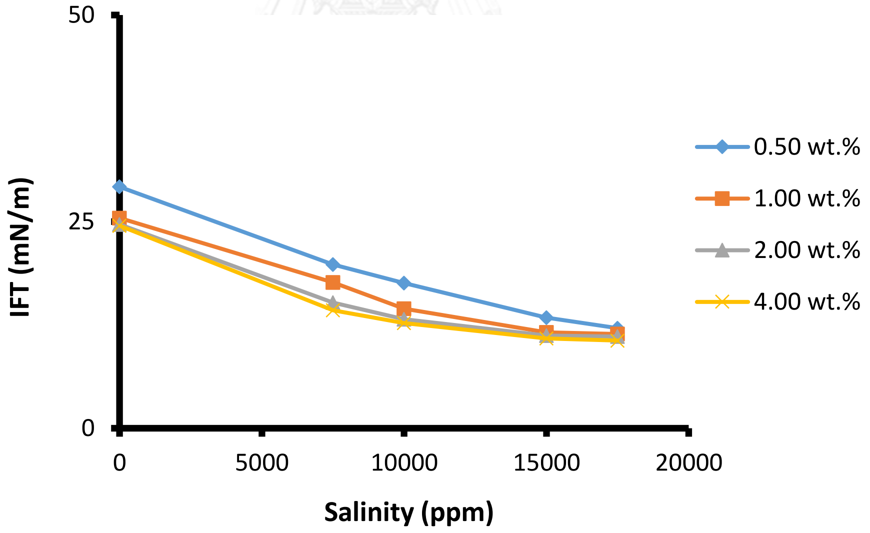

Figure 4.16 Effect of salinity of DEA solution (temperature = 80 °C, pressure = 1,500 psi)

From Figure 4.15 and 4.16, at various surfactant concentrations, the salinity have more impact on IFT when it is compared that without surfactant. The results are shown in Table 4.11. Therefore, the IFT can be lowered when the salinity of brine increases. The reason for this change is that the ions push the molecules of surfactant to oil-water interface. Another reason is that the increment of salinity concentration can enhance the absorption of surfactant at the oil-water interface causing the lowering of IFT (Prosser & Franses, 2001). Besides, at the salinity 15,000 ppm, it can be called the optimal salinity or the intermediate concentration of salt because at this point the IFT is reduced to the lowest value. It means that the amount of surfactant concentration is the same in both oil and water phase and some parts of the surfactant molecules are fully dispersed as monomers at the oil-water interphase

However, at high salinity ranging from 15,000 to 17,500 ppm, the IFT slightly decreases. Particularly, for the case of concentration at 4.00 wt.%, salinity has less significant effect on IFT reduction because high salinity change is to drive the amount of surfactant to the oil-water interface, but the space there is not sufficient for occupying by surfactant. Therefore, some of surfactant molecule that cannot be at oil-water interface will dissolve into the oil phase (Prosser & Franses, 2001).

Table 4.12 The percentage of IFT reduction with increasing of salinity 0 ppm to 15,000 ppm (Surfactant conc. 0.5 wt.%, temperature at 80 °C, pressure 1,500 psi)

Type of solution	% IFT Reduction
Brine without surfactant	2.68
Monoethanolamine	58.49
Diethanolamine	54.23
Fatty Alcohol C1098	19.96
Refined Glycerine	6.05

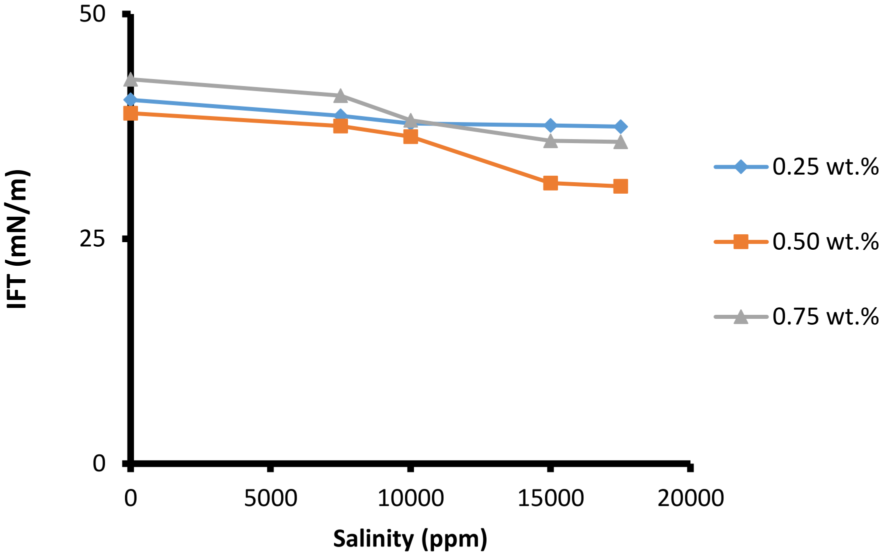

Figure 4.17 Effect of salinity of FAOH C1098 solution (temperature = 80 °C, pressure = 1,500 psi)

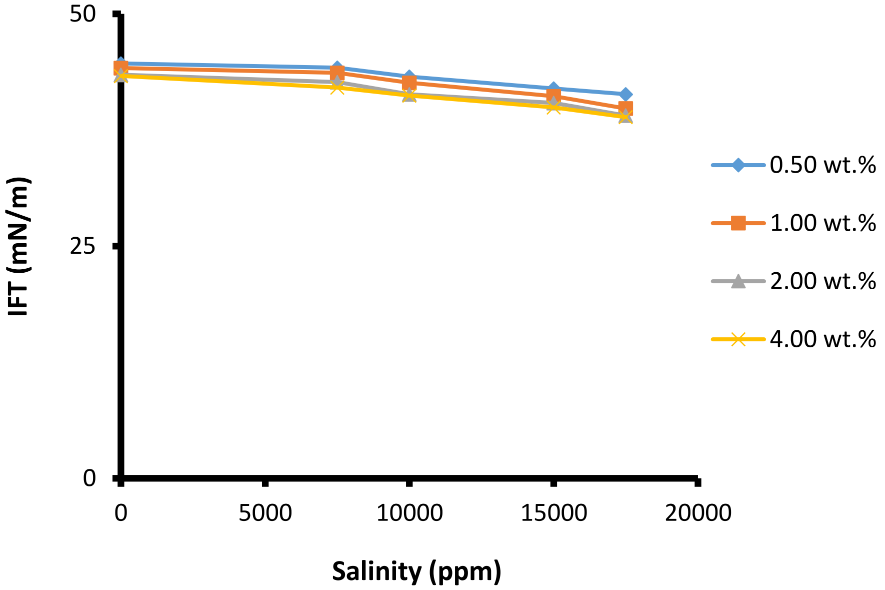

Figure 4.18 Effect of salinity of RG solution (temperature = 80 °C, pressure = 1,500 psi)

The effect of salinity of FAOH and RG are the same manner with ethanolamine solutions. When salinity in the aqueous phase is increased, the interfacial tension becomes lower until it reaches at one point that can make the IFT more stable or become the lowest IFT. However, from Figure 4.17, and 4.18 both fatty alcohol and refined glycerine have less the effect of salinity on IFT although it is added more in surfactant solution due to their ineffective molecules as shown in the Table 4.8 and 4.9 and described in the effect of type of surfactant section.

4.8 The effect of divalent ion on the interfacial tension

As mention earlier in Table 3.3, the divalent ions are not included in simulated brine because they are in a small quantity, 183 ppm for calcium ion (1.07%), and 529 ppm for sulfate ion (3.18%). However, in this section, calcium and sulfate are represented as the divalent ions in this experiment. The effect of divalent ions will be investigated by implementing experiment that is the same with base case condition (MEA 1wt.%, salinity 15,000 ppm, pressure 1,500 psi) and adjusting calcium sulfate to be the same ratio as the produced water from Northern oilfield. The experiment condition varied from 70 °C to 90 °C is conducted to investigate the effect of divalent ions and compared with the results from the base case conditions without divalent ions.

As shown in Figure 4.19, the effect of divalent will increase the interfacial tension. From the theory, the capability of chemical to reduce the IFT can be destroyed by calcium ions (Trujillo, 1983). Accordingly (Agharazi et al., 1990), it confirmed that surfactant can interact with divalent ions forming a high-viscosity, sticky precipitate resulting in decreasing the capability of chemical to reduce the IFT. However, there is less amount of divalent in aqueous phase which is less than one percent mole fraction of the total salinity. Therefore, the results of simulated brine with divalent and without divalent are relatively the same value.

Table 4.13 The effect of divalent from simulated brine

Surfactant concentration	Temperature °C		
	70	80	90
1%wt 15000 ppm (without divalent)	11.27	10.29	9.77
1%wt 15000 ppm (with divalent)	11.72	10.83	9.96

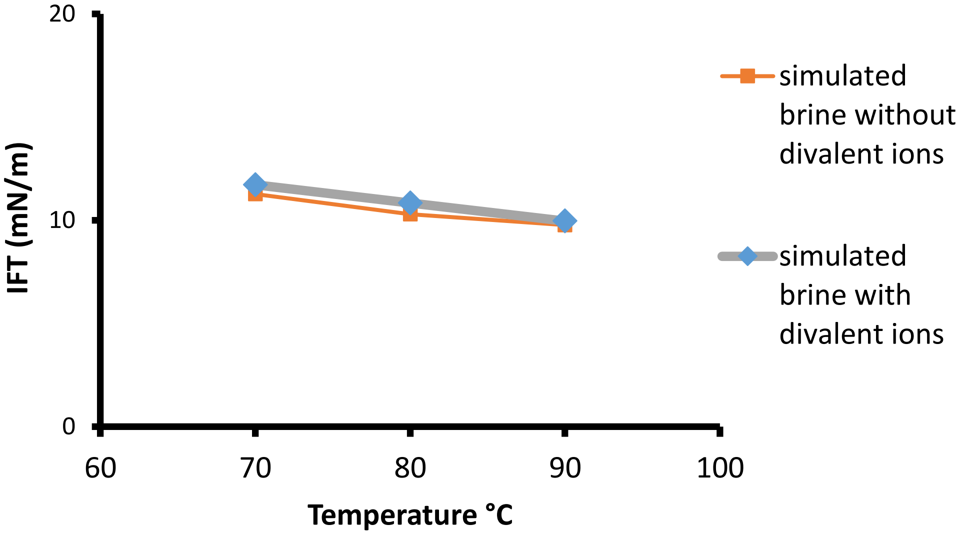

Figure 4.19 Effect of divalent ions on the IFT (MEA concentration = 1 wt.%., salinity = 15,000 ppm)

Table 4.14 The effect of divalent from simulated brine and produced water

Surfactant concentration	Temperature °C		
	70	80	90
2%wt 17500 ppm (without divalent)	5.78	5.46	5.34
2%wt 17500 ppm (with divalent)	5.84	5.74	5.71
2%wt. produced water	8.09	7.93	7.82

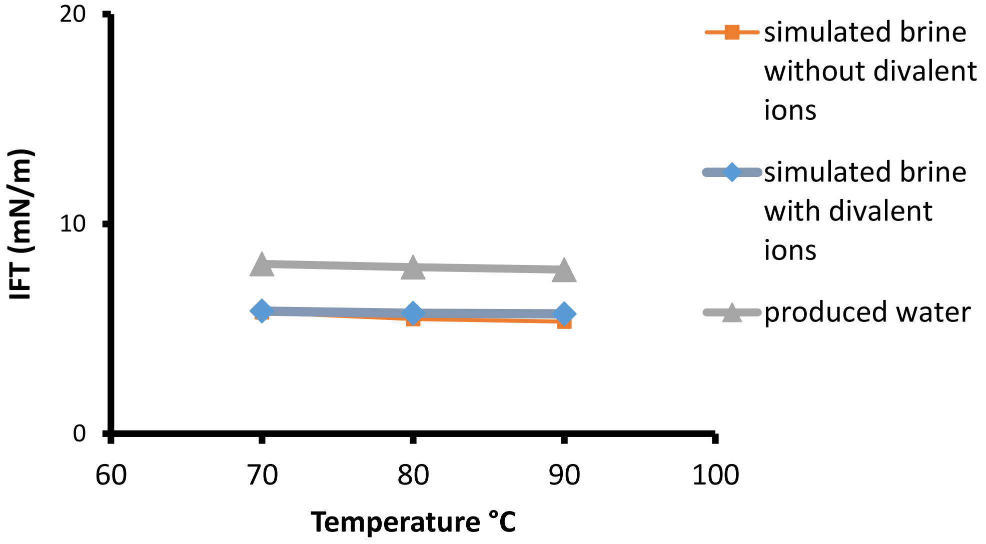

Figure 4.20 Effect of divalent ions on the IFT (MEA concentration = 2 wt.%., salinity = 17,500 ppm

From Figure 4.20, another condition that was implementing was prepared to be the same condition of produced water sample (MEA 2wt.% conc., salinity 17,500 ppm, pressure 1,500 psi) and then the divalent ions was added to observe the effect of divalent ions.

Furthermore, the results from simulated brine with divalent are compared with that of produced water which is obtained from Northern oilfield as shown in Table 4.13. Figure 4.20 shows that the results from produced water has higher IFT value than that of simulated brine with and without divalent ions because produced water contains higher number of suspended solids than that in simulated brine. These could disturb surfactant molecule dispersion on the interface between water and oil more than in case of simulated brine with and without divalent ions. Therefore, it can be concluded that if the surfactant is used on the real conditions, IFT value will be higher than that of from the experiment due to higher suspended solid.

CHAPTER 5

CONCLUSIONS AND RECOMMENDATION

This chapter summarizes all the results from previous chapters including the effects of all parameters on interfacial tension. Moreover, some recommendations are provided in this chapter for the future study.

5.1 Conclusions

In this study, the interfacial tension measurement uses the rising drop method to determine the effects of various parameters such as pressure, temperature, salinity, types of surfactant, and surfactant concentration on the IFT reduction. Also, the advantage of this experiment is to find and screen better effective surfactants that are suitable for decreasing interfacial tension for chemical enhanced oil recovery.

From the results, surfactant solution can reduce the interfacial tension down to 5.46 mN/m for monoethanolamine, 11.09 mN/m for diethanolamine, 30.84 mN/m for fatty alcohol (C1098), and 39.05 mN/m for refined glycerine. However, various parameters that also affect the IFT are investigated during surfactant flooding. The combination of those parameters is required for effective IFT reduction. The effects of each parameter on IFT are concluded as followed

1. The pressure ranging from 1,000 psi to 2,000 psi at constant temperature has less significant effect on IFT reduction. There is small change on IFT when the pressure changes. Therefore, the effect of pressure can be ignored for surfactant flooding in Northern oilfield.

2. Temperature varied in the wide range from 70 to 90°C can always reduce the IFT for every types of surfactant. Although temperature can decrease IFT up to 12.2%, it has less effect on IFT reduction.

3. After screening process based on IFT reduction, there are four different types of surfactant; monoethanolamine, diethanolamine, fatty alcohol C1098, and refined glycerine. All types of surfactant have the effect to lower the interfacial tension; however, from this study, the best type of surfactant which can reduce interfacial tension at the same concentration 0.05wt.% is monoethanolamine. Table 5.1 shows the capability to reduce the IFT of all types of surfactant

Table 5.1 Rank the types of surfactant by their capability of IFT reduction at conc. 0.05wt.% salinity 15,000 ppm, temperature 80°C, pressure 1,500 psi

IFT (mN/m)	% IFT Reduction	Rank
Monoethanolamine	49.76	1
Diethanolamine	47.85	2
Fatty Alcohol C1098	6.57	3
Refined Glycerine	2.55	4
Fatty Alcohol C0898	2.37	5
Fatty Alcohol C1214	1.54	6
Fatty Alcohol C0810	0.69	7

4. The surfactant concentration has a major effect on IFT. For all types of surfactant, the increase in surfactant concentration to certain extent can decrease the IFT. For solution that contains monoethanolamine, the IFT can greatly reduce up to 87.4% as surfactant concentration increases from 0% to 2 wt%, but it is relatively constant at the concentration higher 2.0 wt%. For diethanolamine and refined glycerine, if they are at 2.0 %wt., the IFT can be decreased up to 74.3% and 9.4%, respectively. In addition, the maximum of IFT reduction that fatty alcohol C1098 can reduce IFT 28.4%.

5. The IFT can be lowered up to 61.16% when the salinity increases from 0 ppm to 15,000 ppm for all types of surfactant. However, at high salinity ranging from 15,000 to 17,500 ppm, salinity has less significant effect on IFT reduction. Moreover, this effect would only present when the solution contains surfactant, and it would not much affect if that type of surfactant is not effective surfactant such as fatty alcohol C1098 and refined glycerin.

6. Calcium ion represented as the divalent ions in this study have impact on interfacial tension. However, the divalent ions are in a small quantity in brine composition. Consequently, the effect of divalent ions can be ignored for this study.

7. To reflect the real oilfield condition, IFT value from the real oilfield will be higher than that of simulated brine with and without divalent ions.

8. The interfacial tension can be minimized to the lowest value as following: 1.) Pressure ranging between 1,000 and 2,000 psi, 2.) the temperature at 90 degree Celsius, 3.) monoethanolamine playing a better role than other surfactants on the IFT reduction, 4.) monoethanolamine concentration, diethanolamine concentration, fatty alcohol (C1098) concentration, and refined glycerine concentration at 2.0 %wt., 2.0 %wt., 0.5 %wt., and 2.0 %wt. respectively, 5.) the salinity at 17,500 ppm.

9. These investigated effects of each parameter will be useful to understand the mechanism of IFT reduction, and these results can be used as fundamental data to apply for oil recovery with reservoir conditions

5.2 Recommendation

The following issues are recommended for future study

1. The rising drop method can also measure other types of chemical solution such as alkaline and polymer including mixing of chemical solutions like surfactant-polymer and alkaline-surfactant-polymer.

2. The concentrations that would be investigated had to be varied in wide range because it is very important to observe the point of their CMC.

3. For ethanolamine solutions, they should be investigated the effect of hydrocarbon chain and also compared the IFT value.

4. Surfactants used in this study should be conducted for other applications. For example, adsorption de-inking is a new concept for ink removal from suspension using surfactant solution.

5. The results from this experiment can be used for future studies as fundamental data such as the surfactant core flooding test and simulation.

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APPENDIX

APPENDIX A.

Solution Composition

As shown in Chapter 3, the main chemical composition was sodium ion, and chloride ion corresponding to the produced water composition. Therefore, the brine composition used in this study was Sodium Chloride which would generate sodium ion, chloride ion. The salinity was prepared at 7,500, 10,000, 15,000, and 17,500 ppm. The range of salinity is shown in Table A.1.

Table A.1 Composition of the simulated brine

Salinity (ppm)	Sodium Chloride (g/l)
7,500	7.5
10,000	10
15,000	15
17,500	17.5

APPENDIX B.
EFFECT OF EACH PARAMETER

The effects of each parameter on the IFT are described in Appendix B as described in chapter 4

The Effect of Pressure on IFT

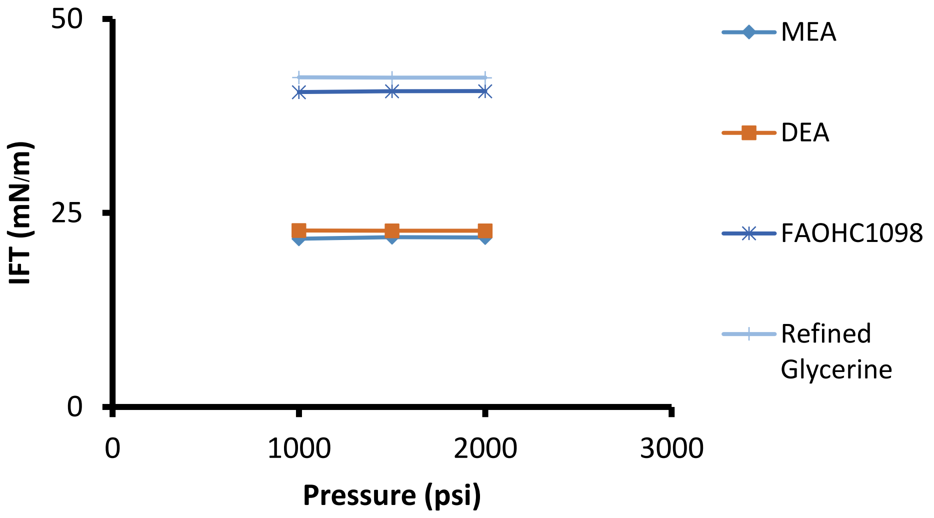

Figure B.1 Surfactant concentration 0.05 wt.%, 15,000 ppm, and 80 °C

The Effect of Temperature on IFT

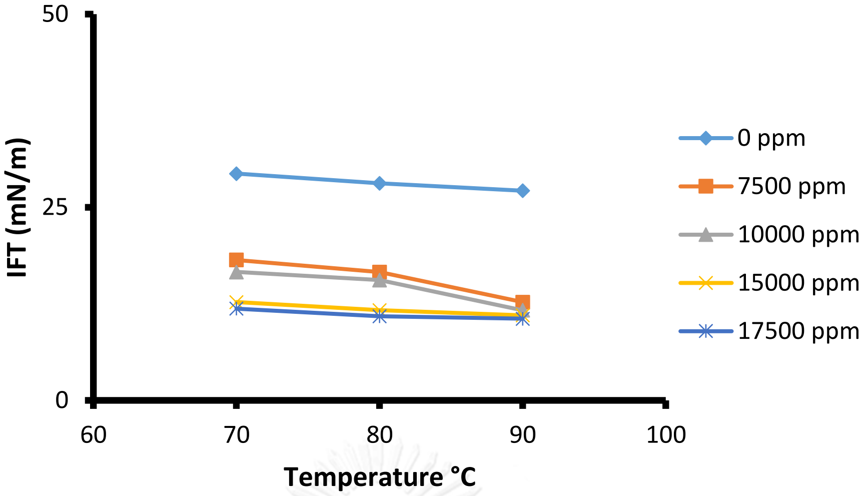

Figure B.2 MEA concentration 0.5 wt.%

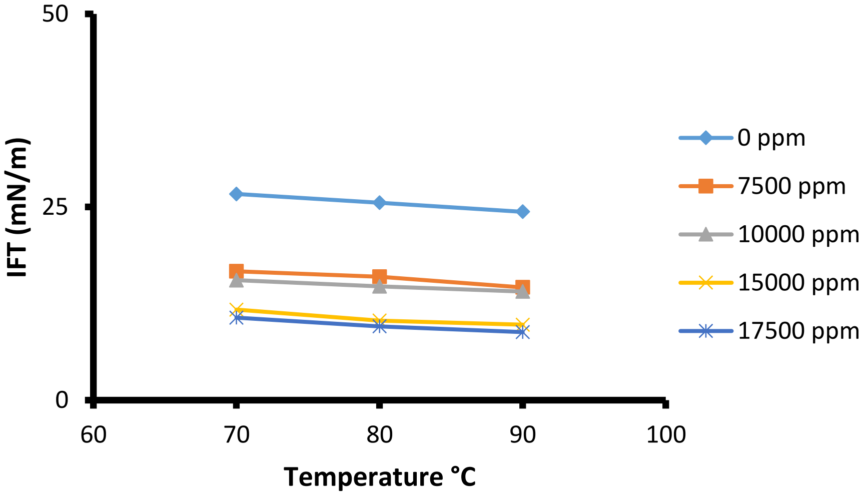

Figure B.3 MEA concentration 1.0 wt.%

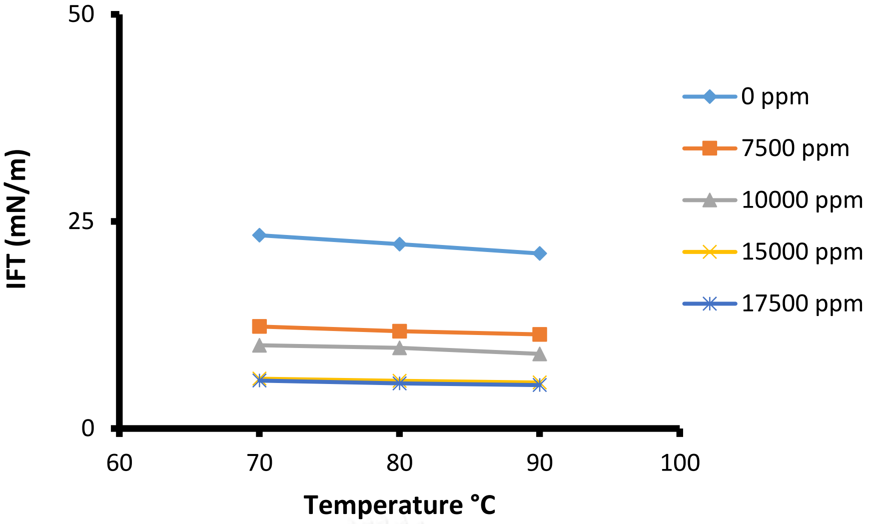

Figure B.4 MEA concentration 2.0 wt.%

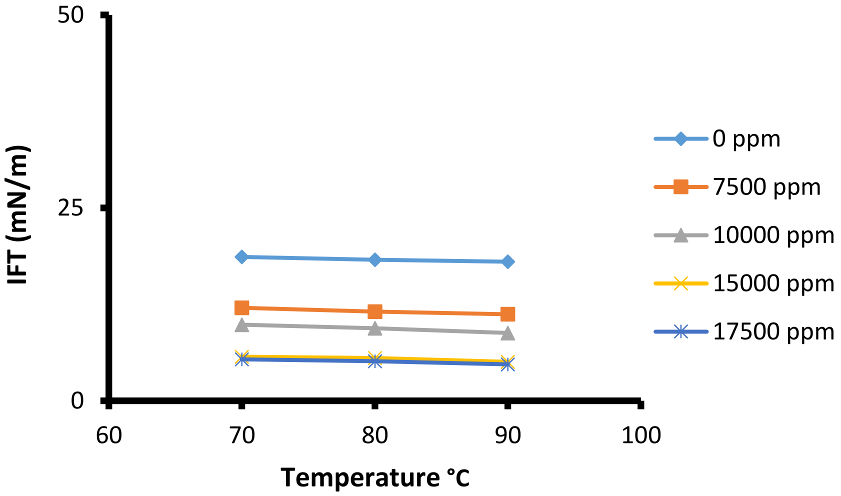

Figure B.5 MEA concentration 4.0 wt.%

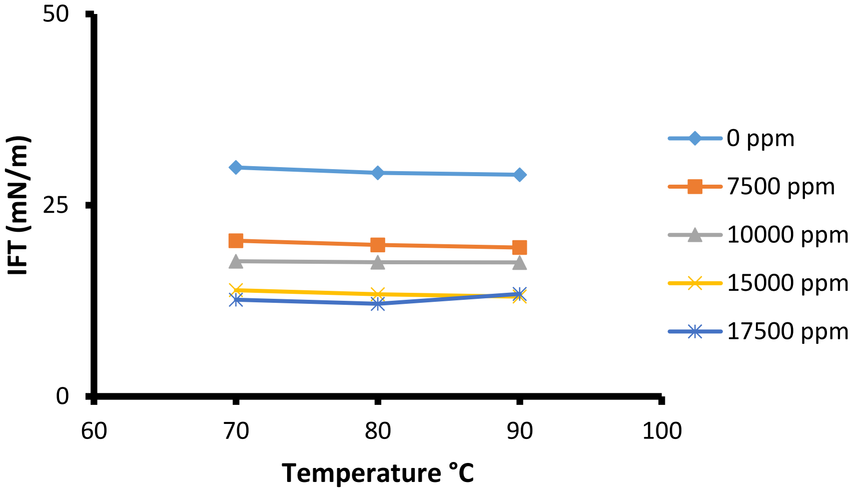

Figure B.6 DEA concentration 0.5 wt.%

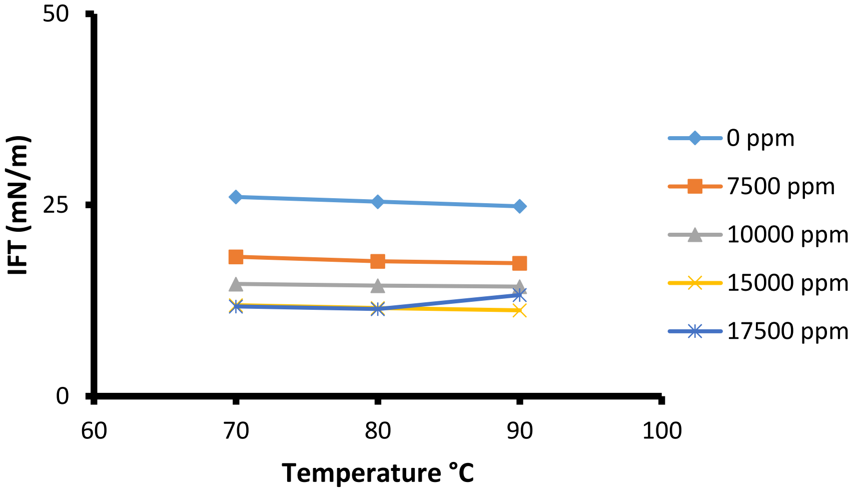

Figure B.7 DEA concentration 1.0 wt.%

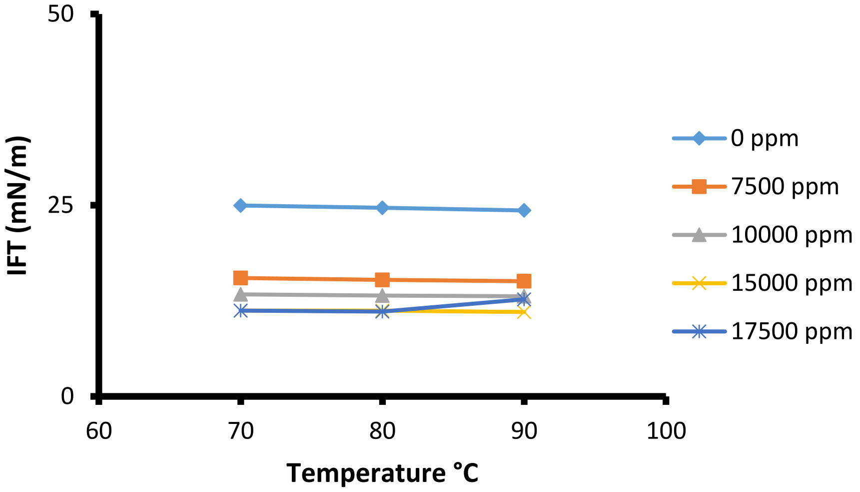

Figure B.8 DEA concentration 2.0 wt.%

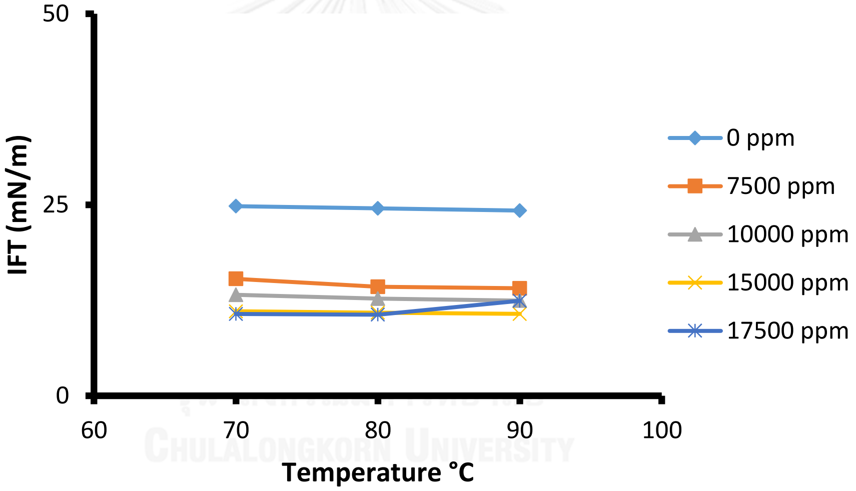

Figure B.9 DEA concentration 4.0 wt.%

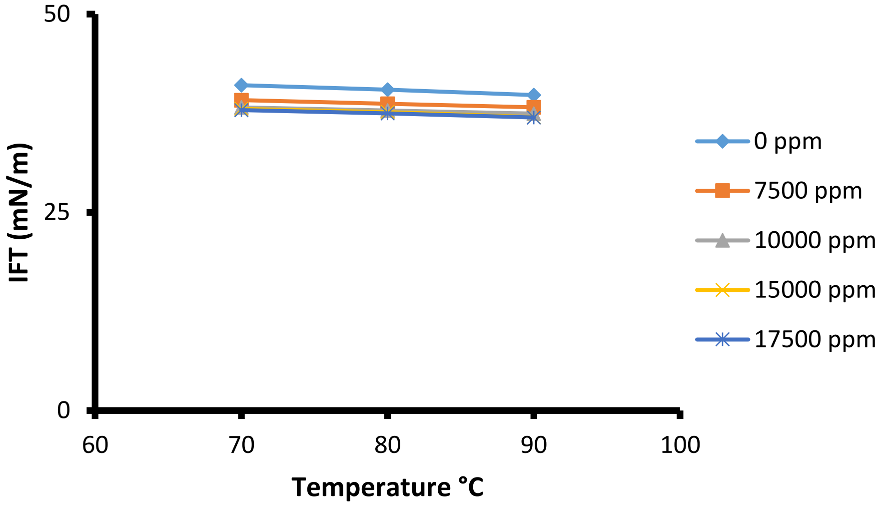

Figure B.10 FAOH C1098 concentration 0.25 wt.%

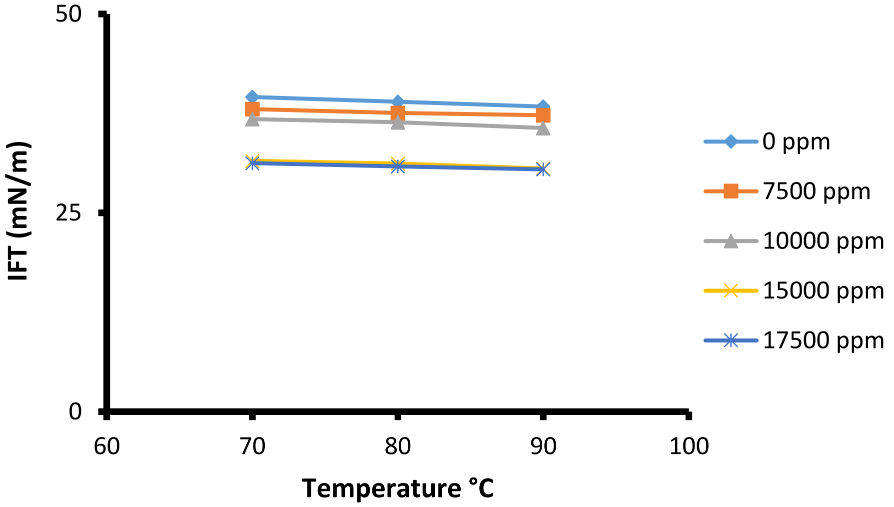

Figure B.11 FAOH C1098 concentration 0.50 wt.%

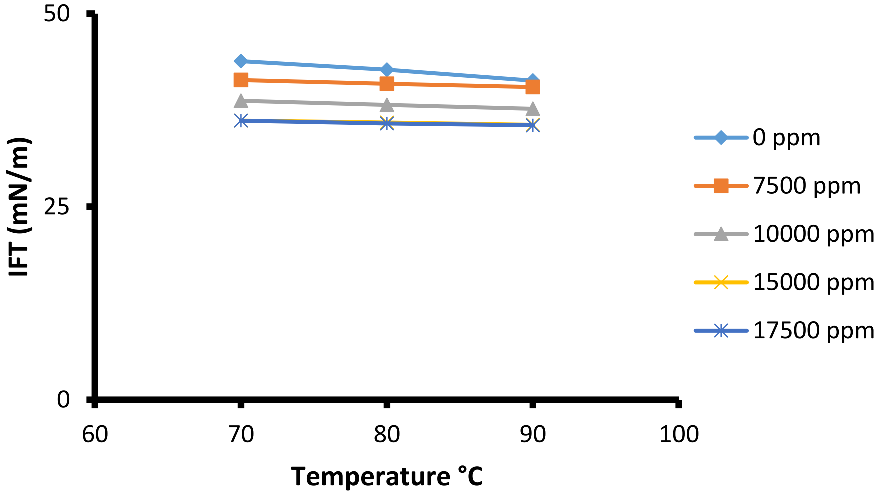

Figure B.12 FAOH C1098 concentration 0.75 wt.%

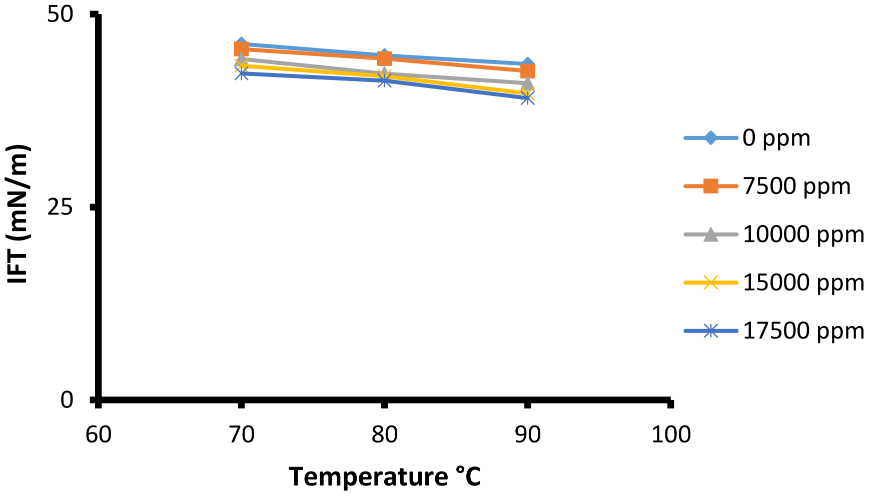

Figure B.13 RG concentration 0.5 wt.%

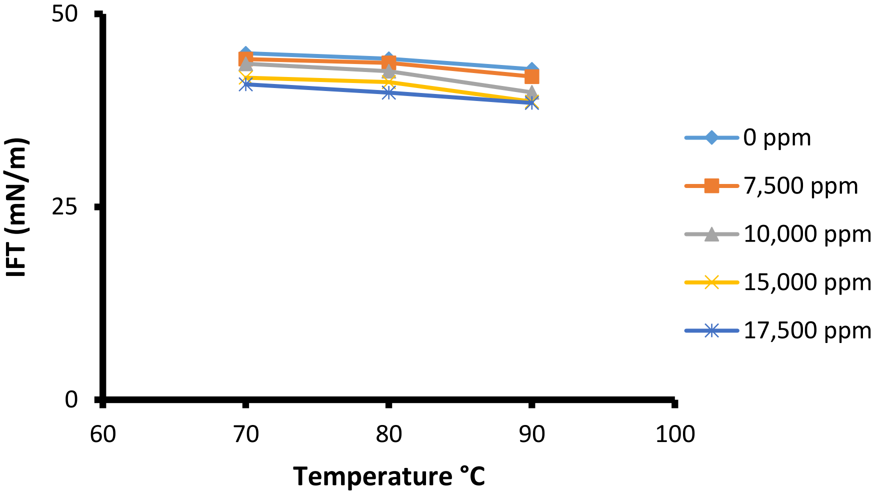

Figure B.14 RG concentration 1.0 wt.%

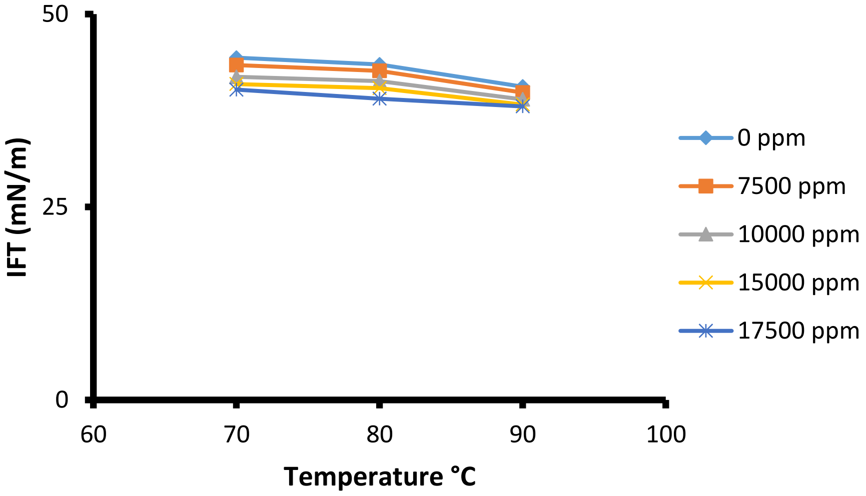

Figure B.15 RG concentration 2.0 wt.%

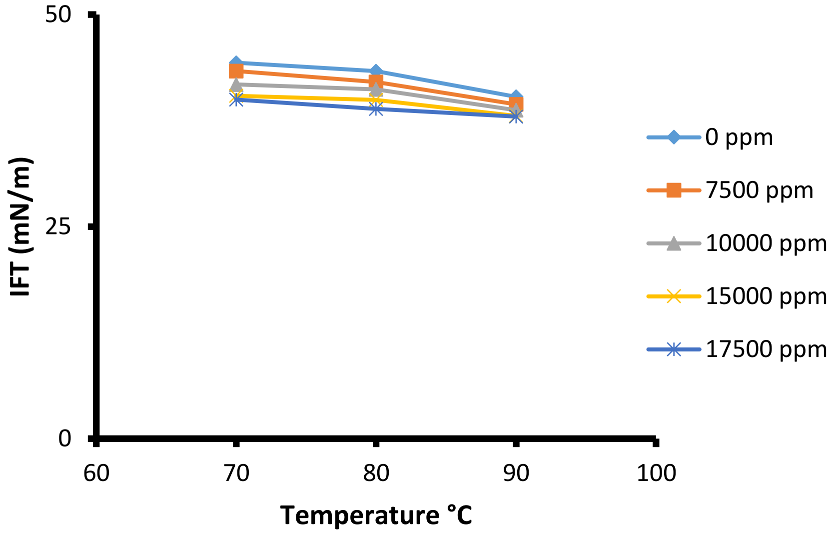

Figure B.16 RG concentration 4.0 wt.%

The Effect of Surfactant Concentration on IFT

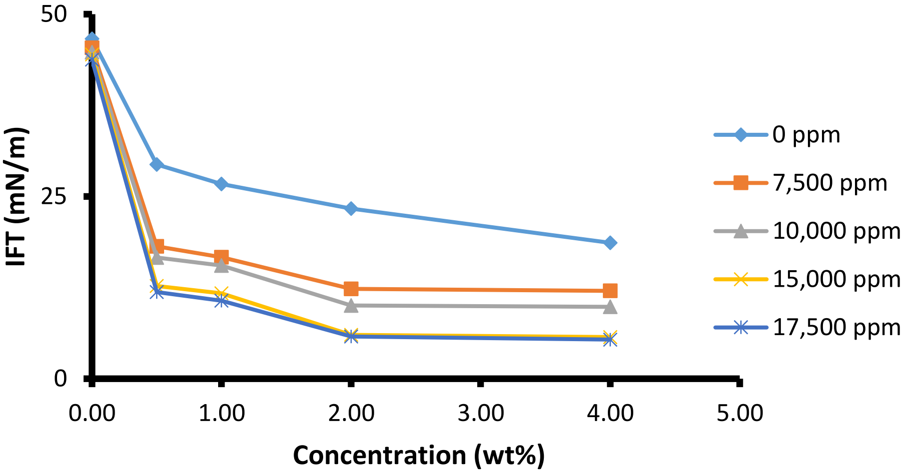

Figure B.17 Temperature 70°C, MEA Solution

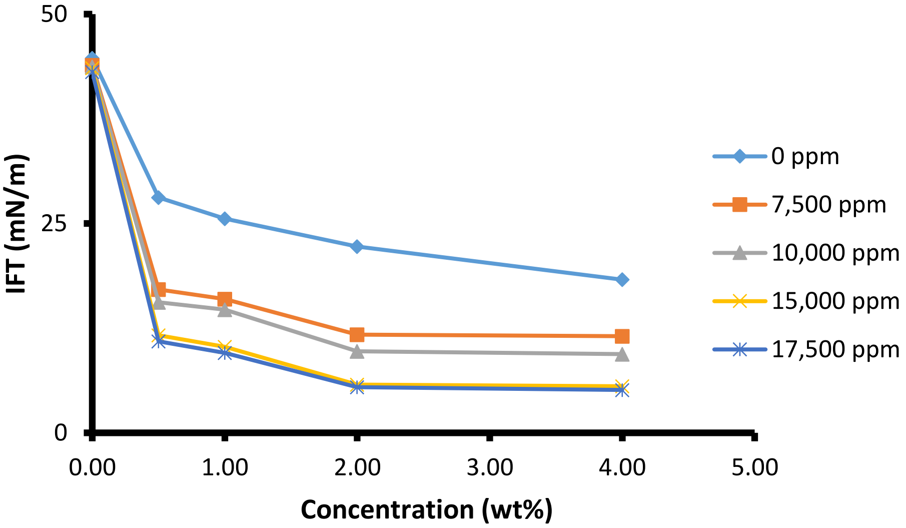

Figure B.18 Temperature 80°C, MEA Solution

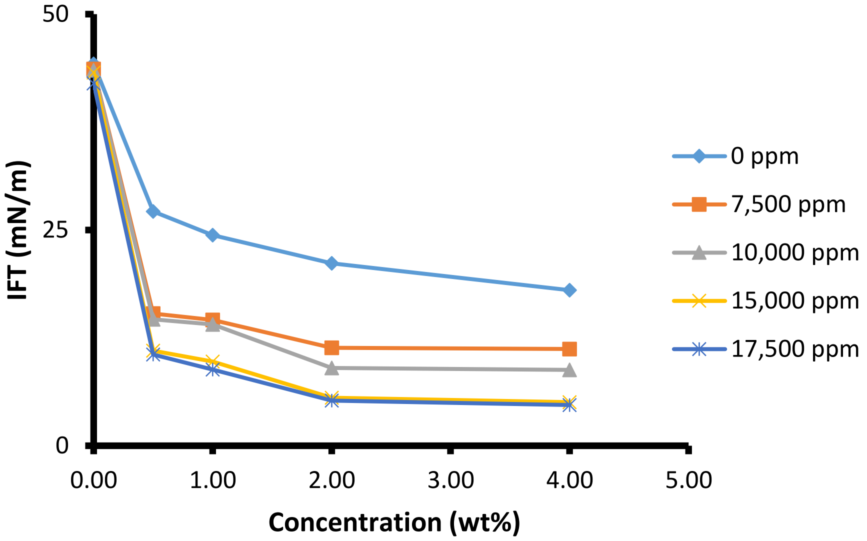

Figure B.19 Temperature 90°C, MEA Solution

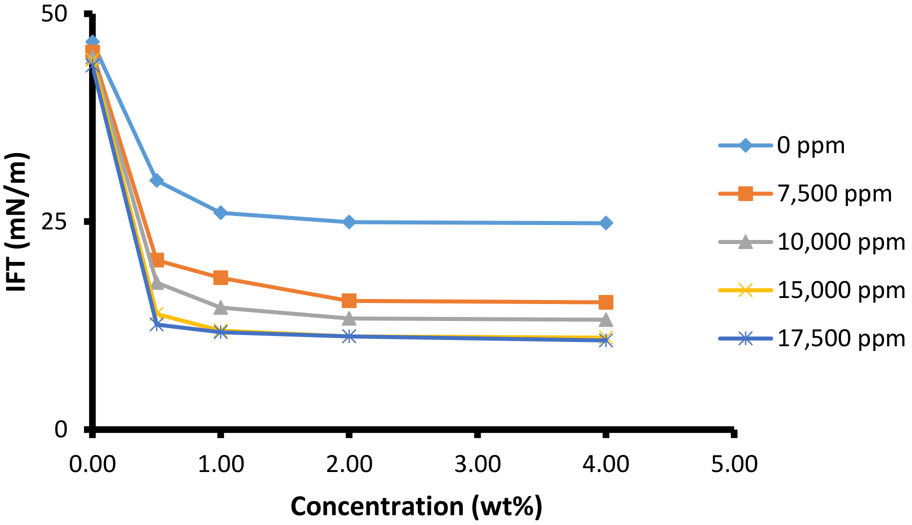

Figure B.20 Temperature 70°C, DEA Solution

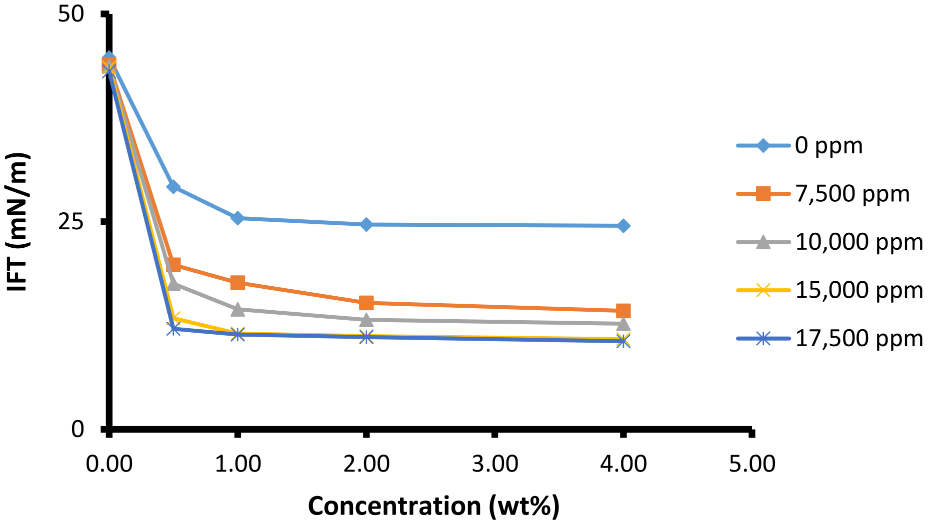

Figure B.21 Temperature 80°C, DEA Solution

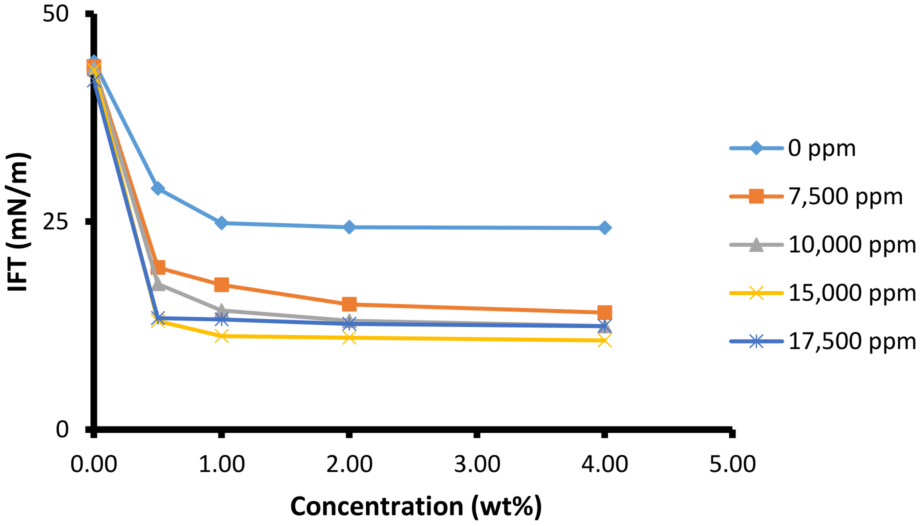

Figure B.22 Temperature 90°C, DEA Solution

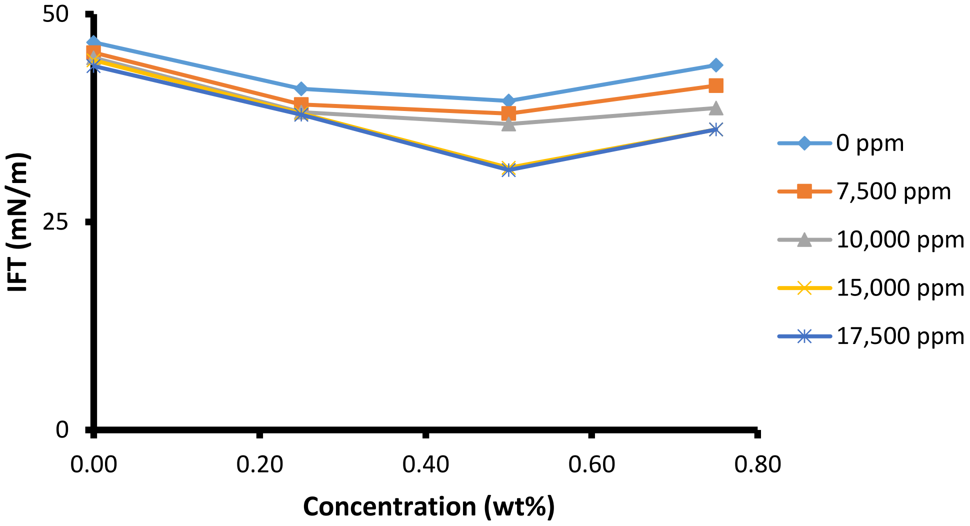

Figure B.23 Temperature 70°C, FAOH C1098 Solution

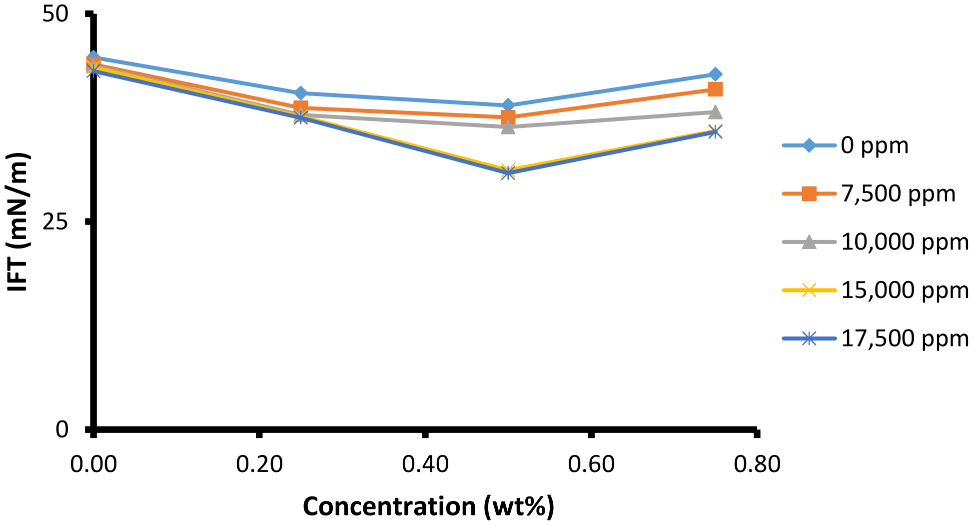

Figure B.24 Temperature 80°C, FAOH C1098 Solution

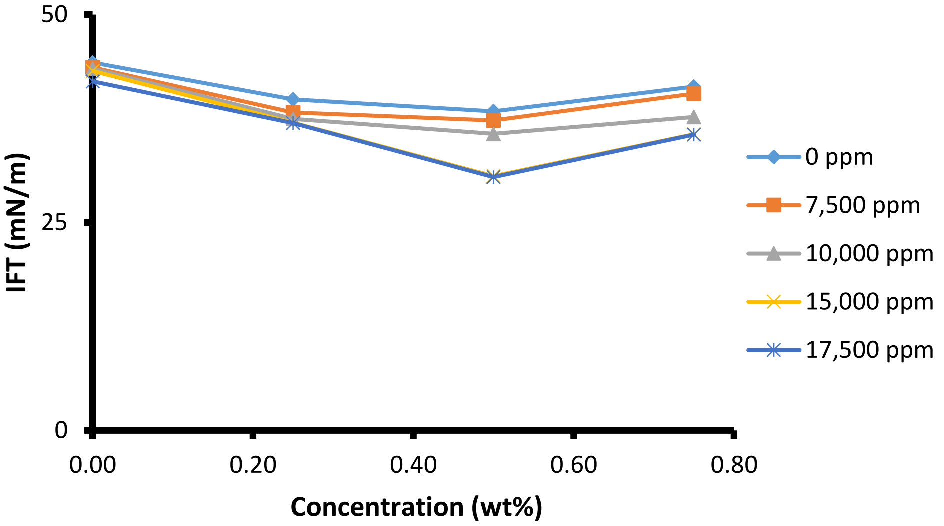

Figure B.25 Temperature 90°C, FAOH C1098 Solution

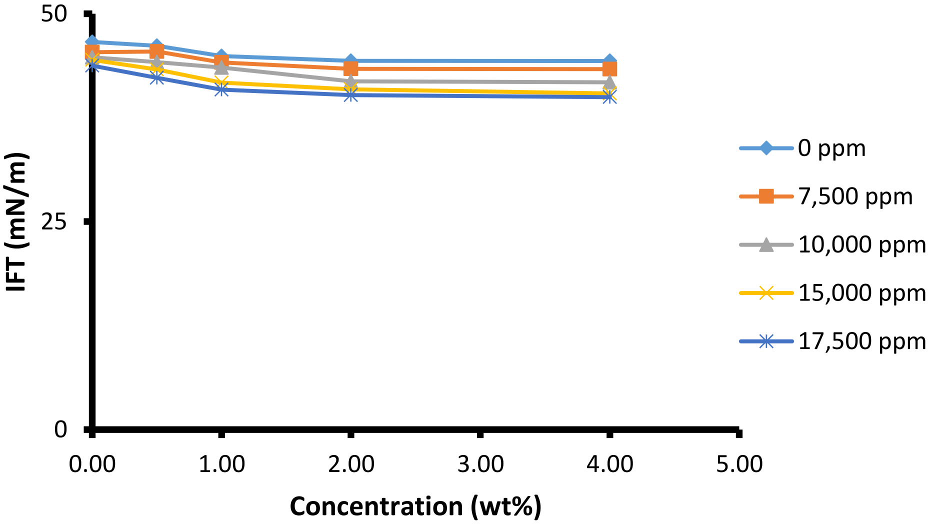

Figure B.26 Temperature 70°C, RG Solution

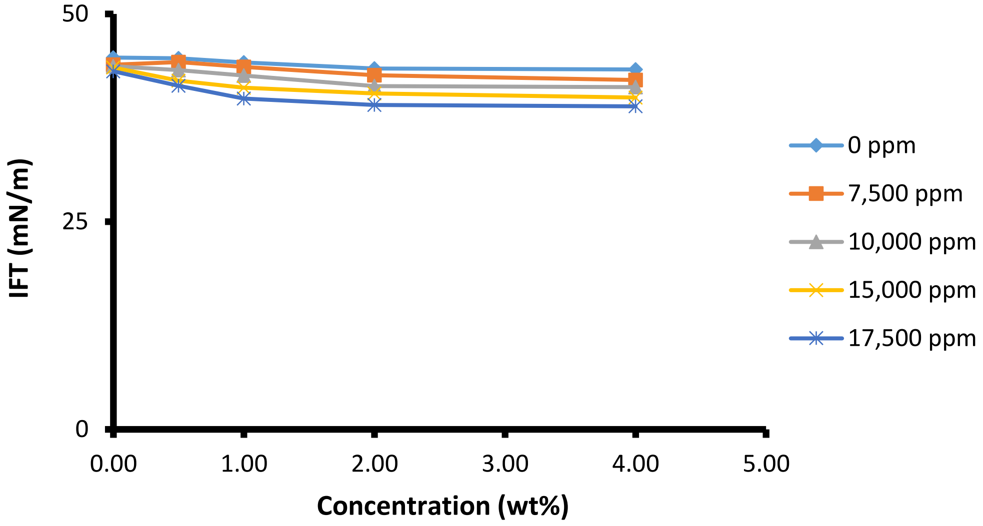

Figure B.27 Temperature 80°C, RG Solution

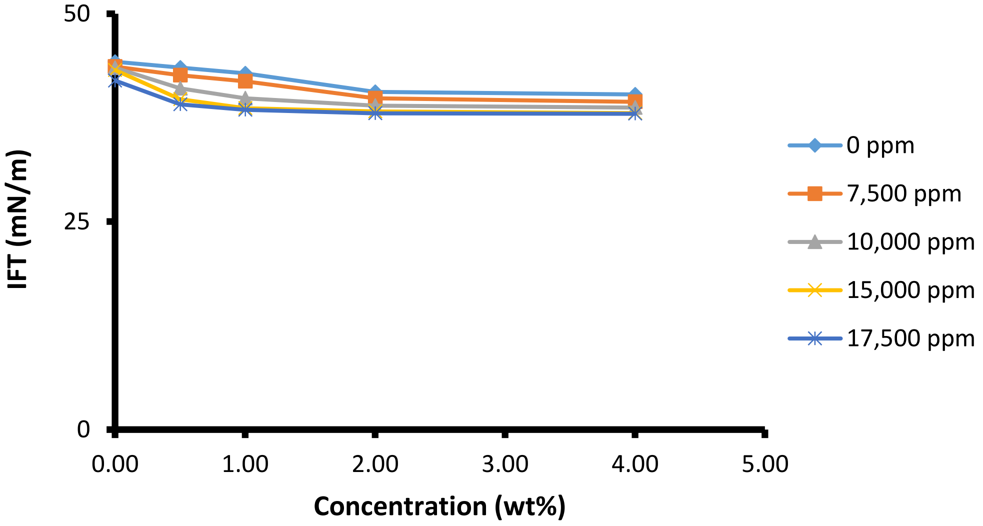

Figure B.28 Temperature 90°C, RG Solution

The effect of salinity on the interfacial tension

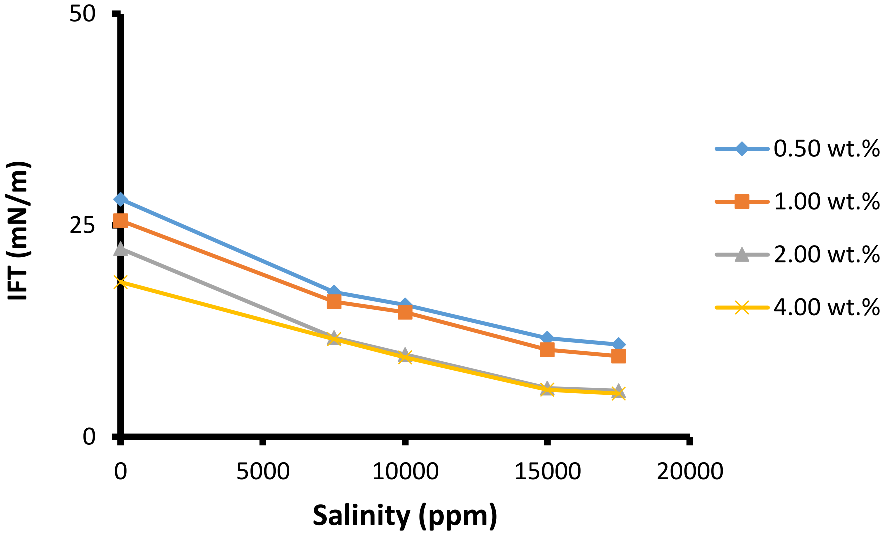

Figure B.29 Temperature 70°C, MEA Solution

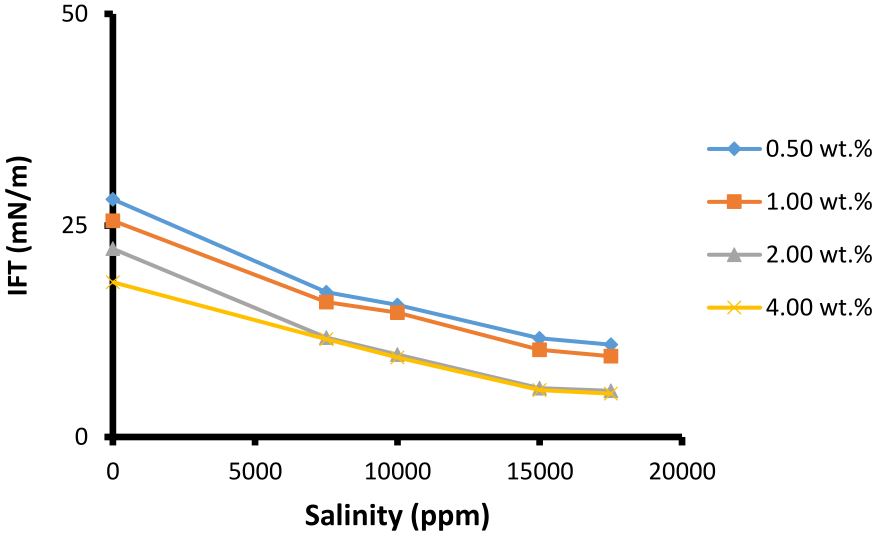

Figure B.30 Temperature 80°C, MEA Solution

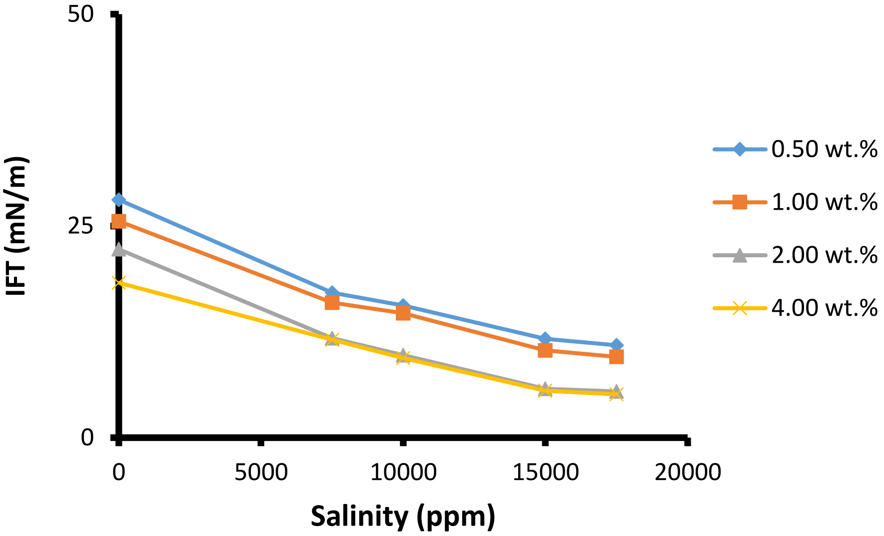

Figure B.31 Temperature 90°C, MEA Solution

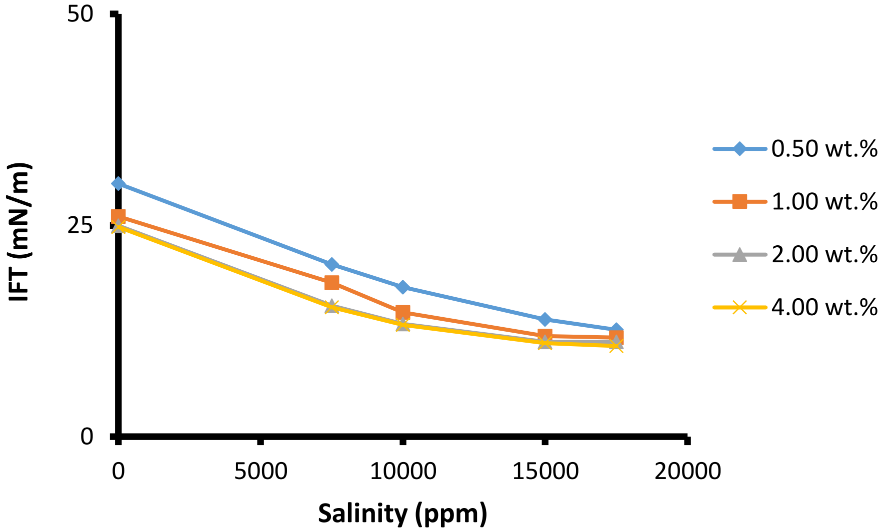

Figure B.32 Temperature 70°C, DEA Solution

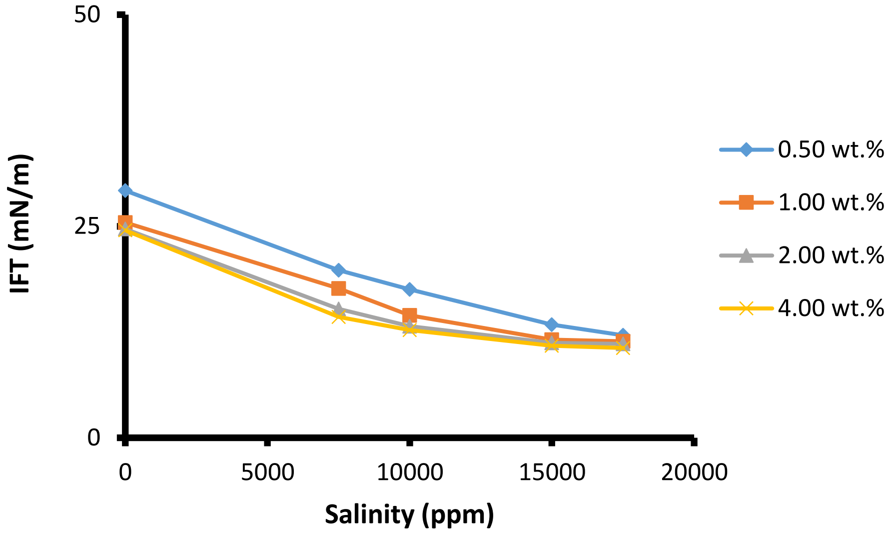

Figure B.33 Temperature 80°C, DEA Solution

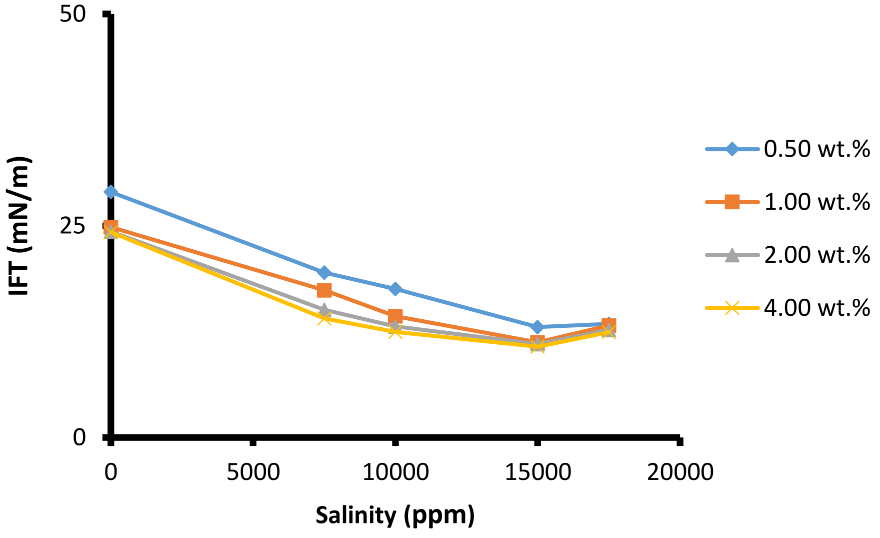

Figure B.34 Temperature 90°C, DEA Solution

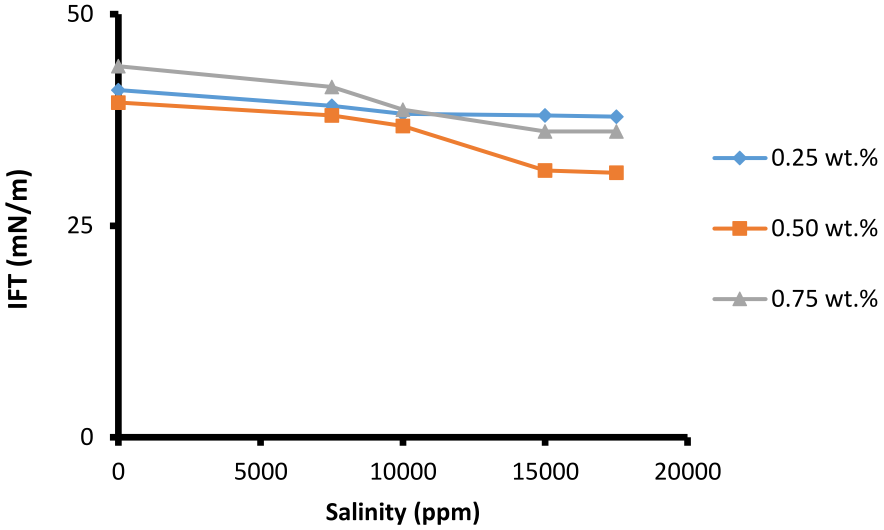

Figure B.35 Temperature 70°C, FAOH C1098 Solution

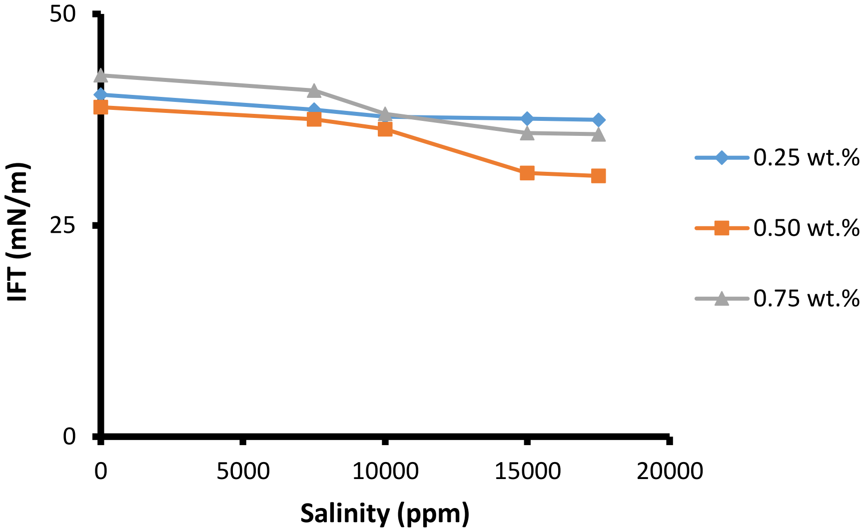

Figure B.36 Temperature 80°C, FAOH C1098 Solution

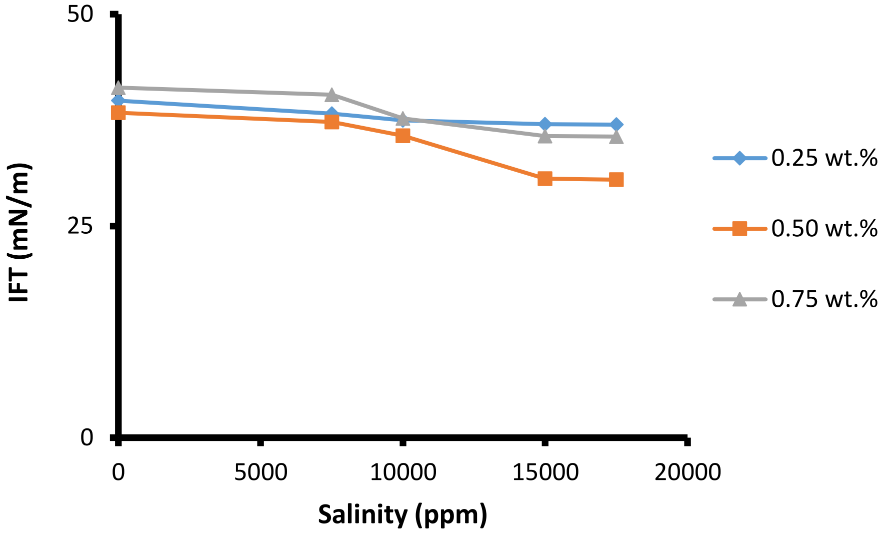

Figure B.37 Temperature 90°C, FAOH C1098 Solution

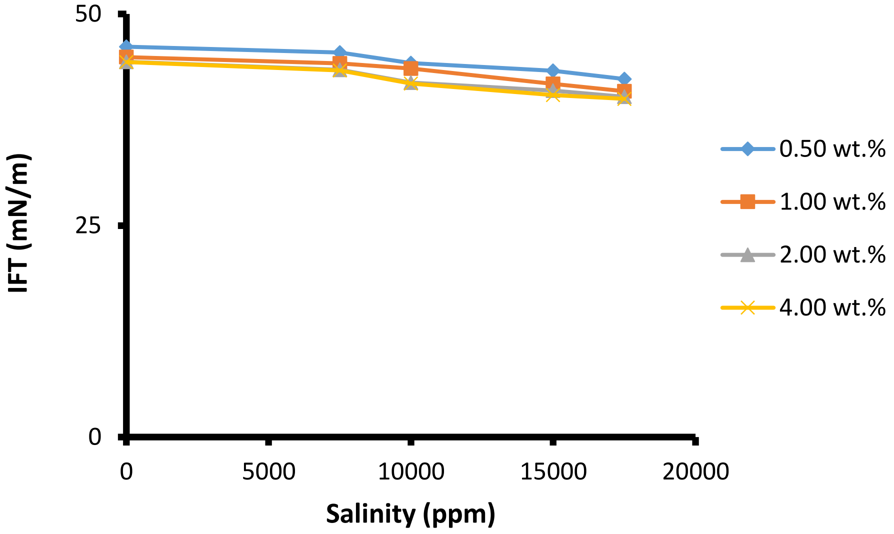

Figure B.38 Temperature 70°C, RG Solution

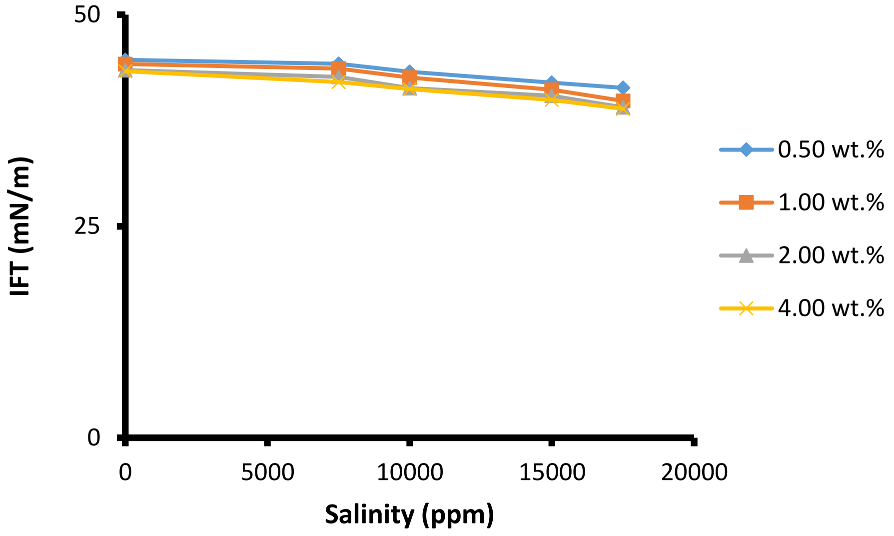

Figure B.39 Temperature 80°C, RG Solution

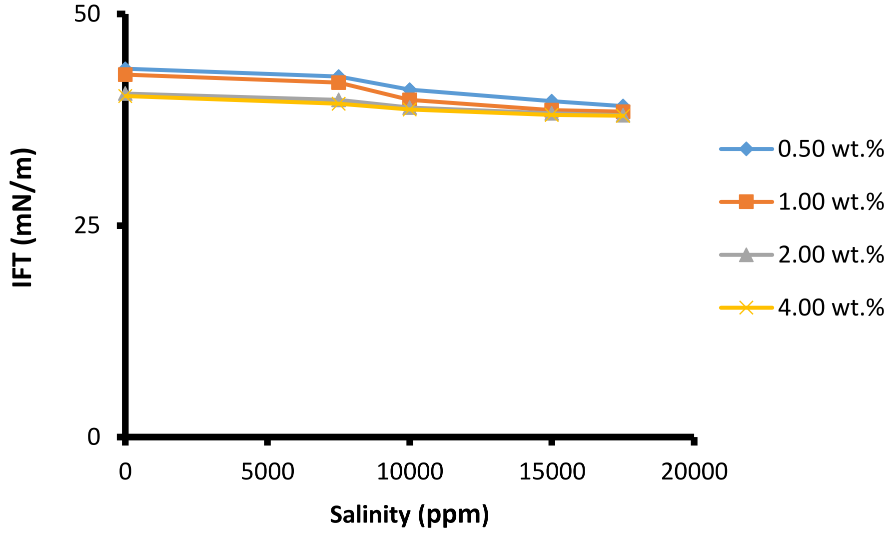

Figure B.40 Temperature 90°C, RG Solution

APPENDIX C.
PERCENT DIFFERENCE TABLE

In order to understand the impact of each parameter more, the results from various conditions are compared with base case condition. Table C.1 to C.6 show the percent difference table.

The base case solutions are 1) Monoethanolamine solution at 1wt.% concentration, 15,000 ppm of salinity, 80°C, 2) Diethanolamine solution at 1wt.% concentration, 15,000 ppm of salinity, 80°C, 3) Fatty Alcohol C1098 solution at 1wt.% concentration, 15,000 ppm of salinity, 80°C, 4) Refined Glycerine solution at 0.25 wt.% concentration, 15,000 ppm of salinity, 80°C

Table C.1 Percent difference of pressure effect on the IFT

Pressure (psi)	1,000	1,500	2,000
Monoethanolamine	-0.9	0	0.1
Diethanolamine	0.1	0	0
Fatty Alcohol C1098	-0.3	0	0.1
Refined Glycerine	0.1	0	0

Table C.2 Percent difference of type of surfactant solution effect on the IFT

Types of solution	% Difference
Brine without surfactant	0
Monoethanolamine	-76.37
Diethanolamine	-73.50
Fatty Alcohol C1098	-.13.59
Refined Glycerine	-5.53

Table C.3 Percent difference of surfactant concentration effect on the IFT

Surfactant concentration (wt.%)	0	0.5	1.0	2.0	4.0
Monoethanolamine	76.37	11.75	0	-43.93	-45.97
Diethanolamine	73.50	13.69	0	-2.86	-5.89
Refined glycerine	5.53	1.95	0	-1.75	-2.92
Surfactant concentration (wt.%)	0	0.25	0.50	0.75	-
Fatty Alcohol C1098	13.59	0	-17.11	-4.54	-

Table C.4 Percent difference of temperature effect on the IFT

Temperature (°C)	70	80	90
Monoethanolamine	12.20	0	-5.05
Diethanolamine	3.02	0	-2.68
Fatty Alcohol C1098	1.08	0	-1.99
Refined Glycerine	1.41	0	-6.10

Table C.5 Percent difference of salinity effect on the IFT

Salinity (ppm)	0	7,500	10,000	15,000	17,500
Monoethanolamine	59.74	35.53	30.05	0	-7.29
Diethanolamine	54.44	34.30	19.79	0	-1.64
Fatty Alcohol C1098	6.97	2.74	0.55	0	-0.40
Refined Glycerine	6.86	5.73	3.36	0	-3.23

VITA

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