ISSN: 2277-3754
ISO 9001:2008 Certified
International Journal of Engineering and Innovative Technology (IJEIT)
Volume 5, Issue 6, December 2015

Influence of Interfacial Tensionon Dynamicsof Multiphase Flow in Mini-Channel

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Abstract*—The dynamics of multiphase flow in mini/micro scale on reaction conversion and mixing in extraction process continues to be a potential research. In mini-fluidic systems, interfacial tension (IFT) forces play a more predominant role in determining flow patterns due to the reduction of channel diameter. Furthermore, the flow pattern strongly depends on the physical properties of fluids and dimensional of the channel. In order to alter flow pattern, the various proportion of alkanes were added into Fish Oil-Silver nitrate binary system and then evaluated interfacial tensionat 25°C using spinning drop densitometry. Additionally, the Bond number for these experimental fluids iscalculated and suggested reasons for flow transition from stratification to slug flow. Even though the addition of alkanes into the fish oil phase does elevate the interfacial tension significantlyand however the stratified flow pattern was observed with Fish Oil–$AgNO_3$ in mini-channel. The reason for stratification is the reduction of interfacial tension at the interface and miscibility between fish oil ethyl ester and silver nitrate solution. By adding sufficient amount of organic solvents into the fish oil phase, the flow patterns in mini-fluidic systems will be tuned from stratified to slug flow.

***Index Terms*— Interfacial tension, Fish Oil, Silver Nitrate Solution, Stratified flow and slug flow.**

I. INTRODUCTION

Due to the reduction of channel dimension, Interfacial tension force plays a major role in the formation of flow patterns. Fish oil ethyl ester–$AgNO_3$ is a rare binary system which lacks information about interfacial tension details and it is evaluated by the spinning drop tensiometry. In addition to that, the effect of the addition of non-polar organic solvents into the fish oil ethyl ester and its interfacial tension was studied. Interfacial tension between organic and aqueous phases plays a major role in extraction experiments. In this process, the larger the interfacial tension, the more readily coalescence of emulsions will occur but the dispersion of one liquid in the other will be more difficult. When the emulsion coalesces more readily, the phase separation might be easier, as the result of high IFT. Low interfacial tension aids dispersion and thus improves contacting mass-transfer efficiency. Coalescence is usually of greater importance, and so the interfacial tension should, therefore, be high. This work investigates about the influence of interfacial tension in fluid dynamics of experimental fluids involved in extraction process at mini scale. [1]

II. MATERIALS AND METHODS

In order to create the slug flow pattern of mini-channel, the interfacial tension properties of fish oil and silver nitrate solution could be elevated by the addition of non-polar organic solvents such as hexane and hexane into the fish oil ethyl ester. To study the flow pattern in Tygon mini-channel, the combination of hexane in fish oil ethyl ester and Hexene in fish oil ethyl ester are prepared.

A. Preparation of Experimental Fluids

The following combinations of experimental fluids are prepared for evaluation of IFT:

1. 100 % Wt. 18/12 EE-Fish Oil Ethyl Esters
2. 10 % Wt. Hexane and 90% Fish Oil ethyl esters
3. 50 % Wt. Hexane and 50% Wt. Fish Oil ethyl esters
4. 10% Wt. Hexene and 90% Wt. Fish Oil ethyl esters
5. 50% Wt. Hexene and 50% Wt. Fish Oil ethyl esters

1) Spinning Drop Tensiometry

The interfacial tension was evaluated for Fish Oil EE–Water, Fish Oil EE–Silver nitrate Solution, Hexane-Fish Oil EE- Silver nitrate solution and Hexene–Fish Oil EE-Silver nitrate Solution at 25°C. The experiment was carried out in Spinning Drop Tensiometer SITE 100 with Drop Shape Analysis Software DSA2. This technique relies on the fact that gravitational acceleration has little effect on the shape of a fluid drop suspended in a liquid, when drop and the liquid is contained in a horizontal tube spun about its longitudinal axis. At low rotational velocities (ώ), the fluid drop will take on an ellipsoidal shape, but when ώ is sufficiently large, it will become cylindrical. Under this latter condition, the radius (r) of the cylindrical drop is determined by the interfacial tension, the density difference between the drop and the surrounding fluid, and the rotational velocity of the drop. As the result, the interfacial tensions are calculated from the Vonnegut equation.

The spinning drop tensiometer SITE 100 is a computer controlled tensiometer for measuring low interfacial tensions of two immiscible liquid -liquid systems. The motor allows rotational speeds up to 15000 RPM (Standard Instrument) or even 20000 RPM. The heavy bulk Phase (Silver Nitrate Solution) is inserted into a capillary which is open to an inlet and an outlet. The drop of the light phase (Fish Oil) is dispensed into the capillary through a septum with a syringe. The measuring cell, inside which the capillary rotates, is thermostatic with oil that serves as lubricant for the rotating parts. However, in the measurement of Interfacial tension with Fish oil EE –Silver nitrate Solution, when rotational speed increases beyond 6000 RPM, the elongation of fish oil ethyl ester breaks. It is

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ISO 9001:2008 Certified
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observed that there is limitation in the measurement of interfacial tension at higher rotational speed.

In addition to that, Fish Oil EE-$AgNO_3$ is a rare binary system which does lack information of fundamental physical properties. The evidence of interfacial tension between fish oil ethyl ester and silver nitrate solution is confirmed by spinning drop tensiometry. As per Vonnegut equation, the interfacial tension between two immiscible fluids are confirmed by plotting the inverse of the cube of the drop radius ($1/r^3$) to the square rotation rate (ω^2), the straight line obtained confirms that interfacial tension between two liquids exists.

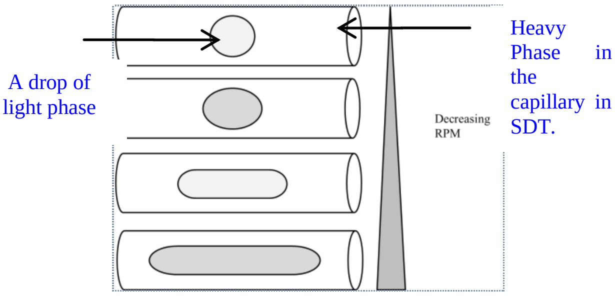

Fig.1. Spinning drop tensiometry.

In this method, the light phase is injected into the heavy phase and forms droplet in the capillary. The drop of fish oil ethyl ester in a narrow capillary tube elongates as the tube is spun along its long axis demonstrating the Vonnegut equation.

Specification of SDT and Operating Conditions:

Measuring Range	Down to 10^{-6} mN/m
Revolution Speed	Up to 15000 RPM
Capillary Diameter	3.5 mm
Temperature range	0-100°C
Magnification	2 microscopic lenses
Power supply	100-240 V

The IFT measured depends on the radius (r) of the drop vertical to the axis of rotation, the circular frequency (ω), and the density difference between the two phases ($\rho H-\rho L$)where ρH and ρL are the densities of the bulk or heavy and light phases, respectively. Therefore, at a given rotational speed with known densities of the two phases and a measured drop diameter, the IFT between the two liquids can be calculated. A condition for accurate measurement is that the length of the drop along the axis of rotation is at least four times the drop diameter ($L\geq 4d$). The above equation is the Vonnegut's equation for calculating the IFT of elongated drops whose length is more than 4 times the diameter [2].

2) Evaluation of Interfacial Tension

The bulk phase (Silver nitrate solution) was injected into the capillary tube of the spinning drop tensiometer, the motor was turned on to run at an rpm of 8500 and the capillary tube was tilted to detect any air bubbles in the tube. Care was taken to remove any air bubble in the tube. A drop of light phase (Fish Oil EE) was then injected into the capillary tube. The rpm was gradually increased and then diameters of light phase are noted. The brightness and contrast were adjusted to get a clear picture of the fish oil EE drop. The temperature was then set at 25°C so as to meet the temperature of extraction. Once the temperature which was set on the control unit was in equilibrium with the temperature of the spinning drop tensiometer and when the drop was long enough to meet the Vonnegut condition, the interfacial tension was measured. Measurements were taken at two minute intervals for 10 minutes. After obtaining the IFT measurements the capillary tube was cleaned with ethanol and hot water before carrying out the experiments with other solutions. [1]

3) Limitations in IFT evaliation

There are certain limitations in measurement of interfacial tension with Fish oil–$AgNO_3$ binary systems. At one extreme the drops are effectively spherical so no measurement is possible and at other extreme the drops are very thin and difficult to measure accurately. In the case of Hexane/Hexene –Fish oil–$AgNO_3$, The droplets are coalesced once the angular speed is increased from 5000. Therefore, it is difficult to measure the IFT at higher angular speed normally > 4000 RPM. [3]

B. Vonnegut Equation and Bond Number.

Therefore, the interfacial tension of the fluid can be obtained using the Vonnegut Formula. This equation is only valid if the ratio of the drop length to diameter *(L/D)* is greater than 4.

$$\gamma = \frac{(\rho_{heavyPhase} - \rho_{lightphase})\omega^2 R^3}{4} \quad (1)$$

The Bond number is defined as the ratio between gravity forces and interfacial tension force.

$$Bo = \frac{\Delta\rho g d^2{}_H}{\sigma} \quad (2)$$

C. Figures

The experimental results are expressed in the figures 2, 3, 4 and 5 and explained.

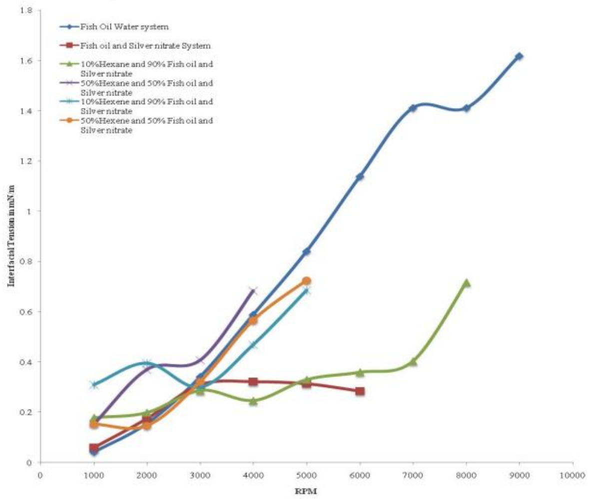

Fig .2. IFT vs. RPM obtained using Spinning drop tensiometry (SDT).

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In this method, interfacial tension is typically the stable value obtained as RPM is increased. Light phase droplet breakup at higher RPMs limited the determination of IFT of experimental fluids.

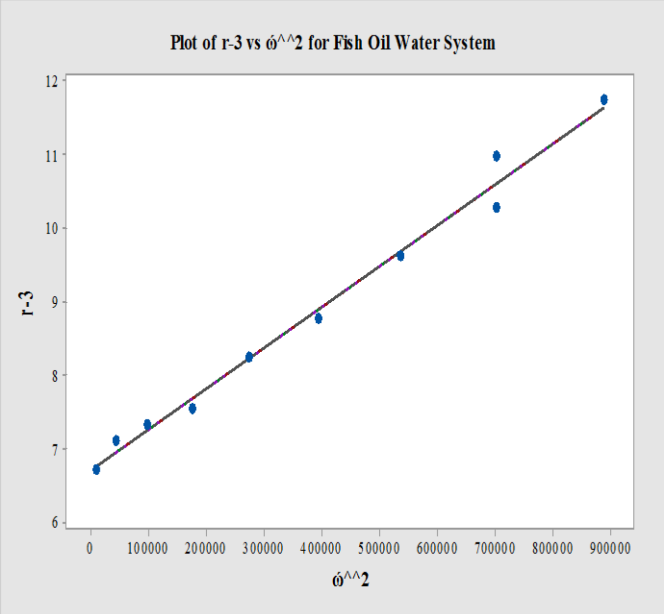

Fig 3A. Fish Oil Water System

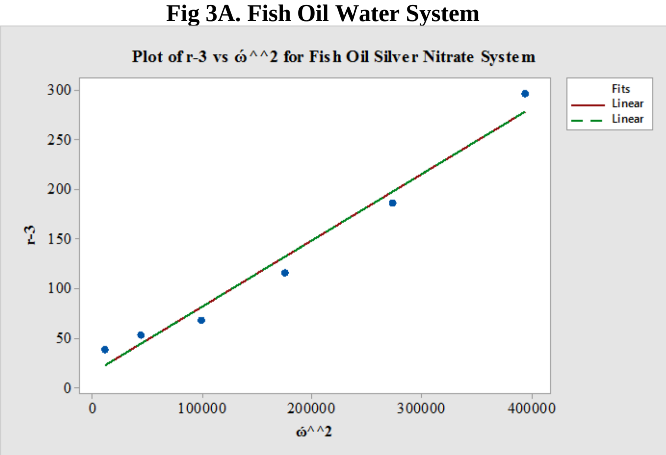

Fig 3B. Fish Oil Silver Nitrate System

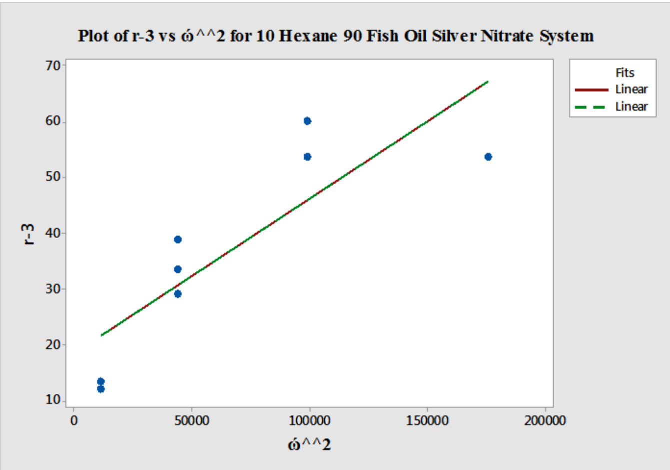

Fig 3C. 10Hexane 90 Fish Oil Silver nitrate System

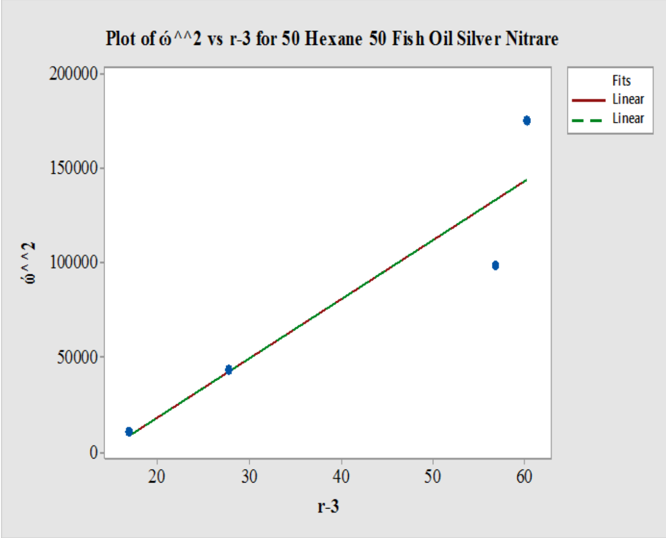

Fig 3D. 50 Hexane 50 Fish Oil Silver Nitrate System

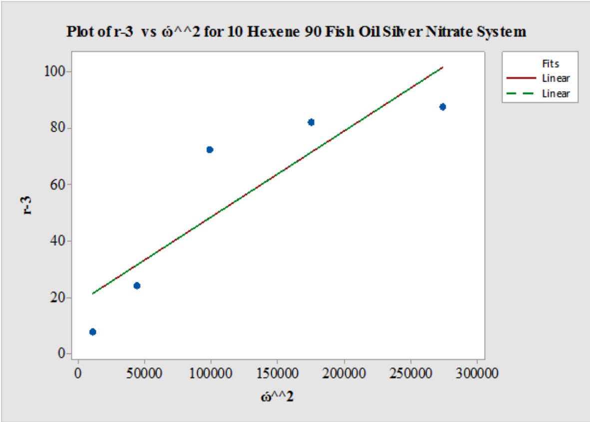

Fig 3E. 10 Hexene 90 Fish Oil Silver Nitrate System

Fig.3. Plot between ω^2 and $r^{-3}$for confirmation for existance of Interfacial tension of experimental fluids such as Fish oil–Water,Fish oil–$AgNO_3$,10% Hexane 90% Fish oil–$AgNO_3$,50% Hexane 50% Fish oil–$AgNO_3$,10% Hexene 90% Fish oil –$AgNO_3$ and 50% Hexane 50% Fish oil–$AgNO_3$.

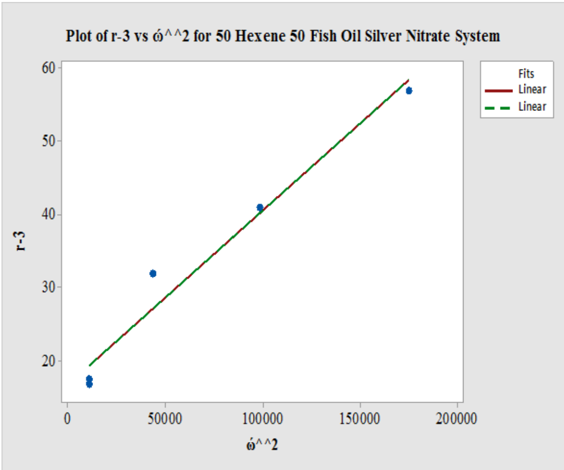

Fig 3F. 50 Hexene 50 Fish Oil Silver Nitrate System

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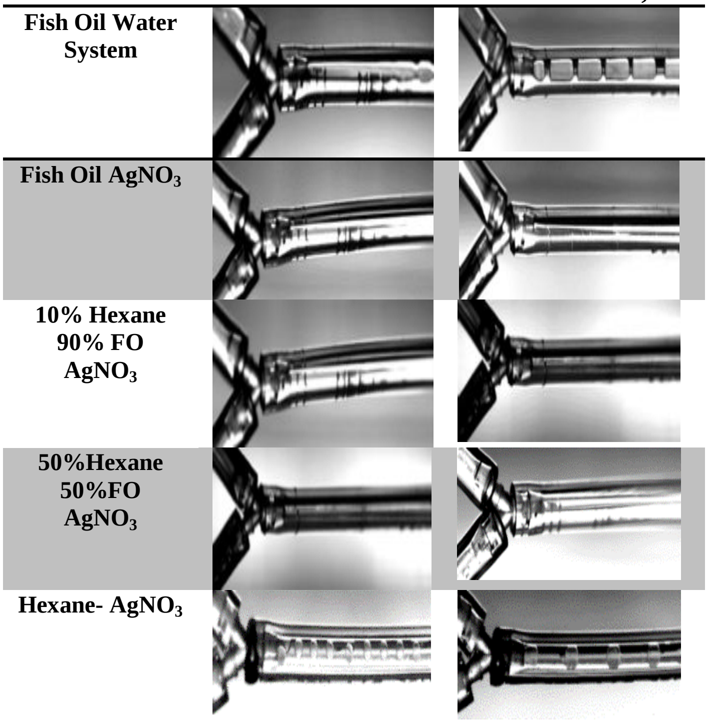

Fig 4. Flow patterns in Tygon mini-fluidic channel. The first row flow rate is 5ml/min of Aqueous phase and second row flow rate is 2 ml/min of aqueous phase. There will be some slug flow formation in the case of Fish oil water system and Hexane Silver nitrate system. But in the case of Fish Oil Silver nitrate system, there is a stratification of flow even alkanes added into this binary system.

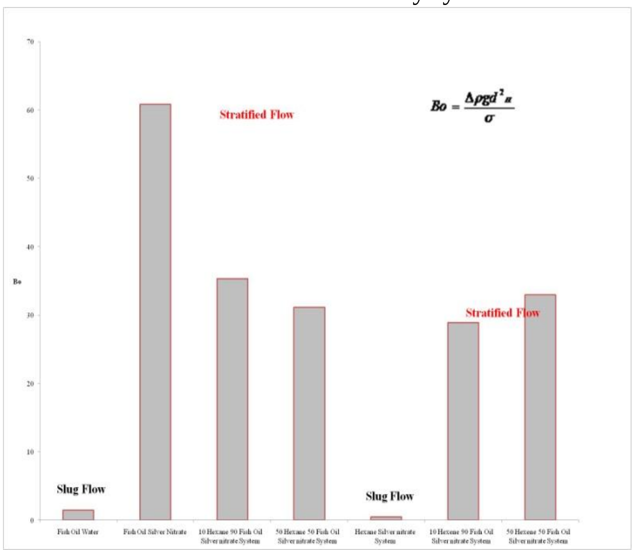

Fig.5.Bond Number (Bo) is plotted versus the different experimental fluids.

The interfacial tension are predicted at 25°C using spinning drop tensiometry. In mini-channel, interfacial forces often dominate gravity forces at Bo<3.37.As a result, the flow is slug/plug flow.Usally, at the mini/microscale, two forces–two forces interfical tension force and gravity dominate with respect to inertial force and viscous force.

III. RESULTS

The data would suggest a significant variation in interfacial tension depending on whether or not silver nitrate is present in the solution, and if hexane and hexene are added to the fish oil, the fish oil and distilled water had interfacial tension which was in the upper limit of the instrument's ability to measure, as illustrated by the steady rise in calculated surface tension with RPM (where RPM's above 7000 to 9000 can create some difficulties in the equipment used). In contrast (Fig .2.), fish oil in silver nitrate solution was at the lower end of the instrument's capabilities, where the interface would breakup up rapidly at rotation rates before stable interfacial tension measurements could be obtained. The 10% wt., of hexane/hexene addition resulted in an apparent increase in interfacial tension. 50% of hexane/hexene had a significant increase in the interfacial tension, rapidly moving past the instrument's ability to measure accurately. However, the addition of hexane either 10 % or 50 % to fish oil with silver nitrate system does not change the flow pattern in the mini-channel and cause only stratified flow pattern with silver nitrate solution. These trends are important for future processing considerations, both in recognizing that the low interfacial tension between fish oil and silver nitrate will facilitate contacting and mass transfer, while possibly making separation difficult. The addition of hexane or, preferably, hexene could use to help in separating the two phases in design of mini-fluidic technology for liquid-liquid extraction of EPA/DHA from fish oil ethyl ester [4].In SDT, Interfacial tension is a function of spinning drop tensiometer rotation. It is noted that the stable value of IFT obtain, as RPM increasing represent the actual interfacial tension[1], [3].In the point of process intensification for extraction process, reduing the dimensions of the channel geometry has an great impact on dominating forces on the fluid flow. In general, the interfacial forces play the most dominant forces especially in two immisible phase flow in minichannel[5], [6].

To compare the gravity with interfacial forces,Bond number is used. However, in multiphase flow in minifluidic channels, gravity forces has no effect for Bo<3.37. Furthemore, in micro/mini-channels, interfacial forces often dominate gravity forces Bo< 1. As can be seen in Fig .5., the interfacial tension results in Bond numbers less than 3.37 only for the fish oil & water and hexane and silver nitrate fluid pairs. As a result, the flow is slug/plug For all other systems, the interfacial tension is too low to give a bond number greater than 3.37 at 1/16" hydraulic diameter of mini-channel[1].

IV. DISCUSSION

Multiphase flows in mini-channel have a wide range of applications in chemical and biochemical processing especially in the case of Liquid- Liquid Extraction (LLE) for omega 3 PUFA from fish oil using silver nitrate solution as solvent. An experimental study of interfacial tension on

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ISO 9001:2008 Certified
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fish oil ethyl ester –water/silver nitrate solution flow in Tygon mini-channel with an ID of 1.58 mm have been performed. Since the Tygon mini-channel was initially saturated with fish oil, the stratified flows were observed. The various flow patterns of fish oil ethyl esters and various aqueous phases in Tygon channel have been visualized. Furthermore, the flow patterns would really affect the extraction processes in food grade slug flow mini-channel.

It is noted that the stratified flow forms in the Tygon channel at the flow rate of 5 ml/min and 2ml/min. The flow pattern map was presented based on Bond number. Two different zones (in Fig .5) were distinguished in flow pattern map namely1. Interfacial tension force dominant–Slug flow region,2. Stratified flow region- gravity force dominant. Furthermore, the addition of organic solvent into the fish oil ethyl ester increase the interfacial tension between fish oil and silver nitrate system, However, There was no transition from stratified flow into slug flow in the fish oil silver nitrate system by addition of hexane into oil phase (Fig 4.). In order to design slug flow based mini-fluidics contacting system for extraction of Omega 3 fatty acids from fish oil ethyl ester using silver nitrate solution, it is important to understand the dynamics of the fish oil silver nitrate system and the addition of organic solvents into the system in the mini-channel. Stratified flow pattern was observed in the mini-fluidic flow of fish oil silver nitrate system.

V. CONCLUSION

The observed flow patterns in the mini-fluidic contacting system were stratified due to reduction of interfacial tension between fish oil and aqueous concentrated silver nitrate solution. However, the formation of oil/aqueous slugs originally were anticipated based on previous literature where LLE performed with DHA/EPA dissolved in organic solvent with silver nitrate solution [4].

Based on qualitative interfacial tension measurements performed on the fish oil/$AgNO_3$ brine mixtures, it is observed that there is a significant reduction in interfacial tension between those fluids relative to a comparable mixture containing quantities of hexane or hexene solvents. In LLE, the low interfacial tension among organic/ aqueous phase would be beneficial to enhancedmass **t**ransfer.It does raise some separation concerns and appears to change the flow regime relative to what has been previously reported in literature for idealized mixtures of purified EPA/DHA in hexane/heptane solvents [4]. Furthermore, in order to improve the performance of mini-fluidics, the study of its hydrodynamic behavior of fish oil silver nitrate system in the channel has been performed. Even though different flow patterns such as slug flow, in some case stratified flow influence mass transfer and mixing [7, 8, 9] which indirectly impact the separation efficiency of omega 3 PUFA from fish oil,the addition of non-polar organic solvents into the fish oil ethyl ester has been attempted to bring the slug flow pattern of fish oil silver nitrate system by elevation of interfacial tension.Furthermore, the role of Bond number in flow pattern formation is commented and justified about stratification of flow in mini-channel.

ACKNOWLEDGEMENT

Kirubanandanis grateful to his supervisor Dr. Adam A. Donaldson for providing funds for graduate studies MASc in Chemical Engineering at Dalhousie University, Halifax, NS, Canada**.**

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ISSN: 2277-3754
ISO 9001:2008 Certified
International Journal of Engineering and Innovative Technology (IJEIT)
Volume 5, Issue 6, December 2015
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