Adiabatic Two – Phase Flow and Flow Boiling in Microchannel

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Lecture - 13
Influence of Operating Parameter on Flow Patterns (Contd.)

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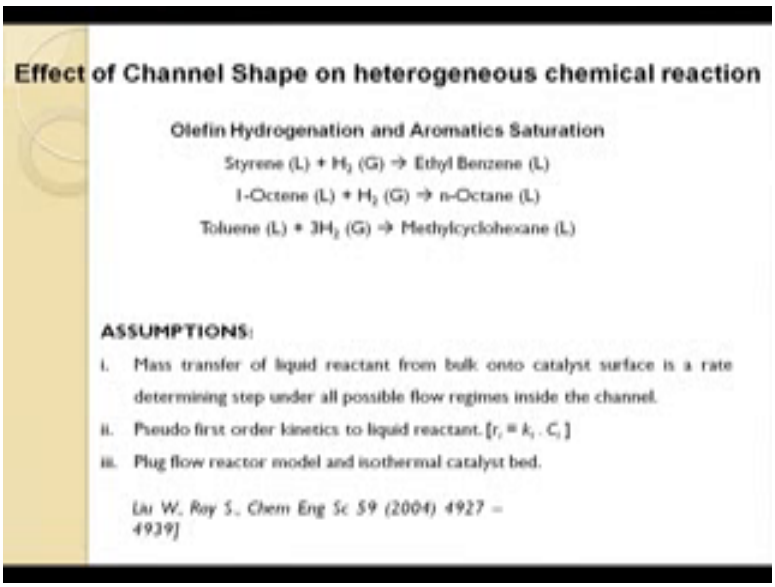

We continue with our discussions today on Gas Liquid Multi Phase Reactions and how they can be enhanced. Therefore, in this respect I would like to tell you that gas liquid multiphase reactions over a solid catalyst, they can be significantly improved by the use of structured instead of fixed bed reactors. Now this all of us know, we know that I fixed bid reactors marginal improvements can be obtained by optimising the catalyst size the catalyst shape gas and liquid distributors catalyst loading method and florizims. People are tried this and they found that that is only marginal improvement in the reaction rate or in the conversion rate, because they fundamentally do not fundamentally change the way the gas and liquid are reacting or rather did not fundamentally it effect they make gas liquid and the catalyst they contact each other.

Therefore, instead of fixed bed catalyst, people have then shifted to structure catalysts. Now this structure catalysts what we mean is that these catalyst they are actually characterized with the straight flow channel and the flat catalyst surface. Which represents and this as represented a dramatic change, over the conventional pellet

catalyst where randomness is very much prevalent. And this structure catalyst they also have some other advantages, not very delivent multiple flow they provide but support and also they provide confinement to the 2 phase flow and they have been very successfully used in automotive catalyst converters and they are very commonly termed as monolith catalyst or honeycomb shaped, honeycomb reactors on monolith reactors. Now in this particular reactor, what we have actually is, we have a honeycomb shaped structure where a large number of channels are drawn and though this channels the reactant flow and reaction occurs where the wall of the channel serves to be the catalyst and therefore the gas liquid reactions occurred there.

Now, these automotive catalyst converters while they are very popular for gas phase catalyst reactions, people have found that they could not be use with much success for gas liquid catalytic react conversions. And the primary reason was because it was difficult to engineer the gas liquid flow morphology which would maximise conversion. So, naturally lot of work was being is being persuaded I should say on how to increase the conversion in this particular monolith reaction for heterogeneous solid catalyst reactions.

In this respect good amount of work has been done, but relevant to a lecture is the study by Liu and Roy, where they have adopted 3 particular hydrogenation reactions and till these 3 hydrogenation reactions this styrene hydrogenation, I octane hydrogenation and toluene hydrogenation. Out of these the first 2 are kinetically very fast, while the last one it is kinetically slow.

Also we are seen that in the first 2 reactions they obey or rather that they can very well represented by the language type of kinetic situation and therefore if we can insert a dilute feat stream the reaction kinetics it can be approximated by first order kinetic reaction. This was the result for selecting these reactions and what the researches they date was, they performed this particular reaction in square as well as circular micro channels. It is not exactly micro they are used 2 mille meter and to one mille meter dimension and that studied 2 things, one was the influence of channel wall thickness where they found that the channel the thickness of the channel wall did not significantly influence the conversion at the shape of the channel because mostly in monolith reactor since it is easier to make square or rectangular channel. So, that was being preferred.

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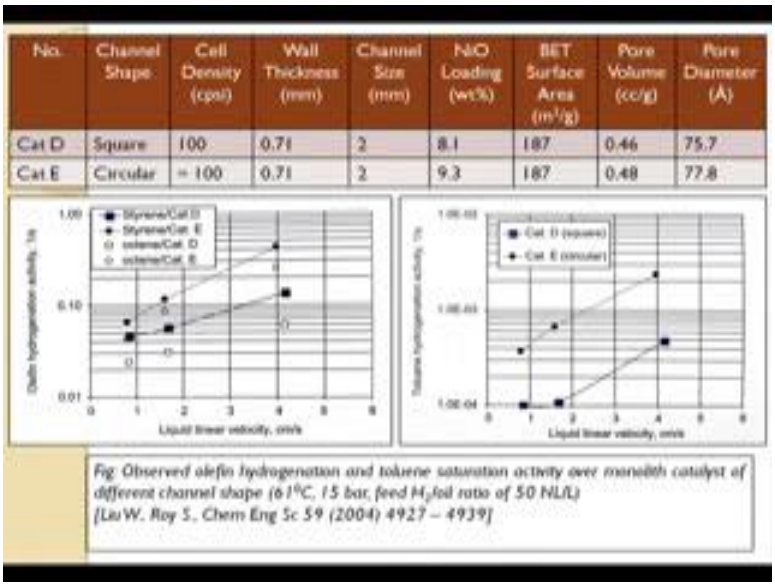

Fig: Observed olefin hydrogenation and toluene saturation activity over monolith catalyst of different channel shape (61°C, 15 bar, feed H_2/oil ratio of 50 NL/L)
[Liu W. Roy S., Chem Eng Sc 59 (2004) 4927 – 4939]

They try to see the influence of conduit shape on conversion. And this is the result which they have observed. They had used 2 particular catalyst, catalyst d and catalyst e as it is represented and they have in this particular 2 graphs, they have shown the conversion of only if in, as well as the conversion toluene this is a fast reaction this is a slow reaction and it is interesting to see that. Firstly, for both the cases we see that the conversion rate or the activity it increases with linear velocity as expected this happens for all the cases.

Secondly, we find that the conversion rate is much higher for the case of a circular as compare to a square channel and the increase of the enhancement in activity for toluene hydrogenation is about 2 to 3 times whereas, for toluene hydrogenation which is the slow reaction I would like to remind you the increase in activities about 3 to 5 times.

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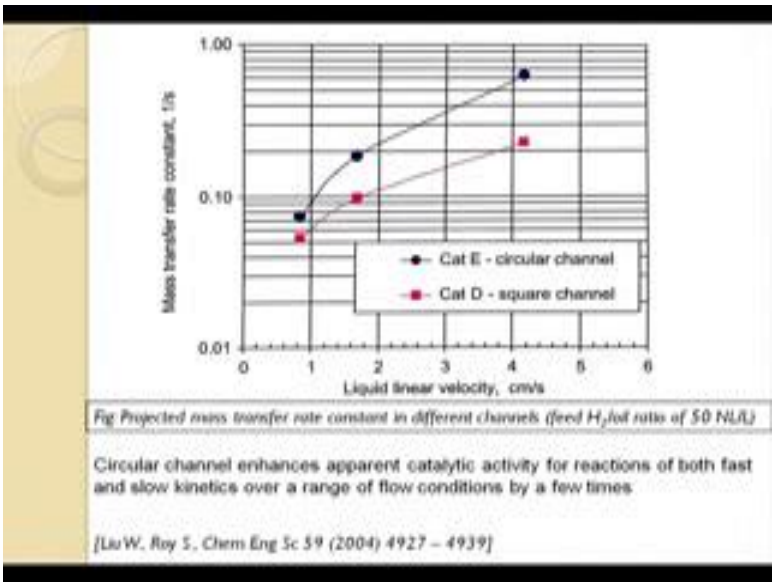

Which shows that circular channel enhances the apparent kinetic activity for reactions of both fast and slow kinetics, over a range of flow conditions not only at few, but over a range of flow conditions by a few times. Well after that and the same thing has been shown by the mass transfer constant as well. In this particular case also we see that the mass transfer constant increases and for all velocities and definitely for the circulate channel the mass transfer constant is much higher.

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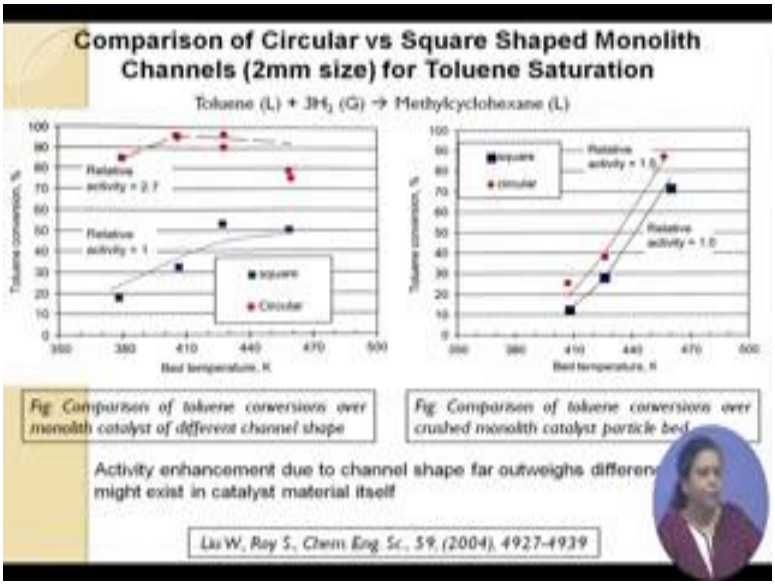

Than peoples expected that whether this activity enhancement was due to some particular difference in the catalyst material itself, whether if the way they were they were made, whether that was affecting the catalyst activity.

In order to check it what they did. Firstly they try to find whether it was really happening for both ico octane as well as toluene. What they did? They prepare rather they try to just con confine the reaction to toluene hydrogenation. For which they added small fractions of toluene into methyl cyclohexane, in order to ensure that only toluene saturation was occurring and then they perform the reaction in both a circular and square conduit.

For just toluene conversion in the absence of any other reaction also the same thing happened and the enhancement in reaction was rather the circular channel was a much better the reaction activity as compare to a square channel. Well then they wanted to see weather really there is something to do with the catalyst material rather than the catalyst shape. For that, what they did? They took the catalyst, they crush the catalyst, they sewed it in to 100 to 200 mesh of particles. Therefore, when the crushed it the shape factor is no longer there and then they tested the activity in a trickle bed reactor configuration with silicon carbide as the diluents.

Therefore, here they were testing with the circular and square monolith reactors and in this case with the case of crushed catalysts. Now when they tested the activity with the crushed catalyst they found that, in this case also the circular monolith reactor had a slightly increased activity as compare to the square monolith reactor, but the relative activity in this particular case was just 1.5, whereas when the shapes were retained then in that case the increases activity was 2.7.

Therefore, from this it is very evident that the activity enhancement due to the channel shape it far out ways, the differences that might exist in the catalyst material itself and therefore the differences which might exist due to catalyst impregnation their physical properties etcetera that does not affect the catalyst activity as much as the shape effects. And then they try to investigate that why is such a situation occurs? And they found out that it primarily occurs because of the type of wetting or the wetting characteristics of a circular and the square channel. They found out that for proper reaction to occur the surface needs to be covered completely by a liquid film and gas should be diffusing in

this particular film. Only when gas and liquid both the reaction are in contact with the catalyst, only then the reaction occurs and the faster rate of reaction occurs.

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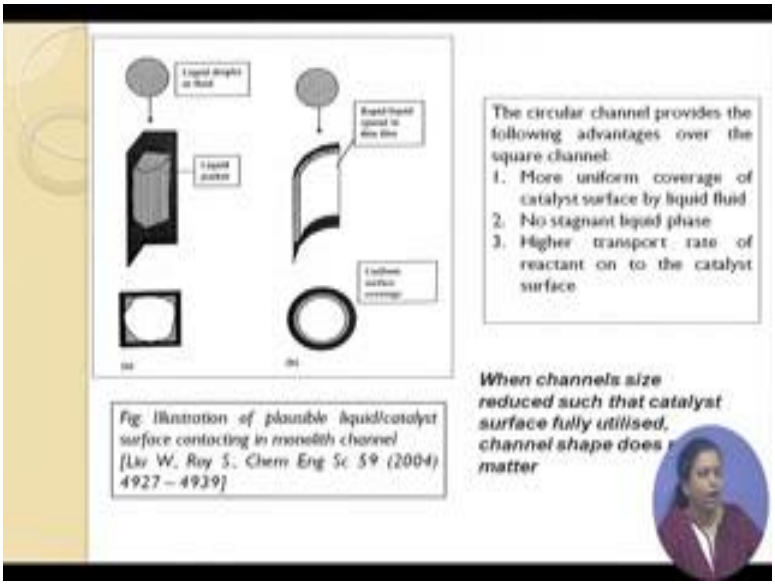

Now, for the circular to be find, that there is more uniform coverage of the catalyst surface by the liquid film and there are no stagnant liquid phases, there no sharp edges or corners where the liquid can get trapped and remains stationary, the gas cannot approach that region and those regions are not available for reaction.

On the other hand if we observe the square catalyst, what we find is that definitely this particular case also the square catalyst also if we observe it can be deemed as a morphologically smooth surface. Well and it can also be enclosed or rather it can also be leered or it can also have thin film on it, but due to the corner effect if you find we find that all though it its morphologically smooth just like the circular passage, but it has a large number of stagnant of the death zones, which preferentially tend to trap the liquid and these types are not available for reaction.

Therefore, the catalyst activity in order to a get enhanced conversion, it is not only important that the it forms a very thin film and it ensures gas liquid reaction, but it is very important that both gas and liquid they can approach the catalyst surface and the reaction can occurred. Interestingly this entire discussion which I had was for catalyst dimension of 2 mille meters. The circular conduits were of 2 milli meter diameter and to the square conduit 2 milli meters of edge. Now when the catalyst size was reduced or at

the conduit dimension was reduced to 1 milli meter we found that defiantly the the reaction was enhanced the catalyst activity was defiantly enhanced, but the enhancement for the case of square conduit was much more as compare to the enhancement is the conduit of circular conduit.

What does this explain? This explain that in a circular conduit in the 2 milli meter case also moral list it was ensuring efficient contact of gas and liquid with the catalyst surface. Further reduction in dimension does not enable a larger amount of liquid-liquid gas solid contact. Whereas for a square this surface was inefficiently utilised in 2 milli meter and a reduction in size guaranteed or enable better utilisation of the catalyst surface and a greater enhancement in the read. From this it was also concluded that suppose we keep on reducing the channel size till a point when the entire channel surface gets properly utilised.

Now after that further reduction in catalyst size, will not bring about any further enhancement in the conversion of a reactor. So therefore, the catalyst size or rather the catalyst surface it matters only to the extent till the catalyst cannot be fully utilised, once fully utilised the channel shape does it not matter, but in this particular miniaturised systems, in this in this particular range tin he range in which the monolith reactors generally operate the range between see 0.5 milli meters to 3 milli meters this is the range were they generally operate. In this particular range the channel shape definitely matters a lot as per this studies which have been performed by Liu and Roy well.

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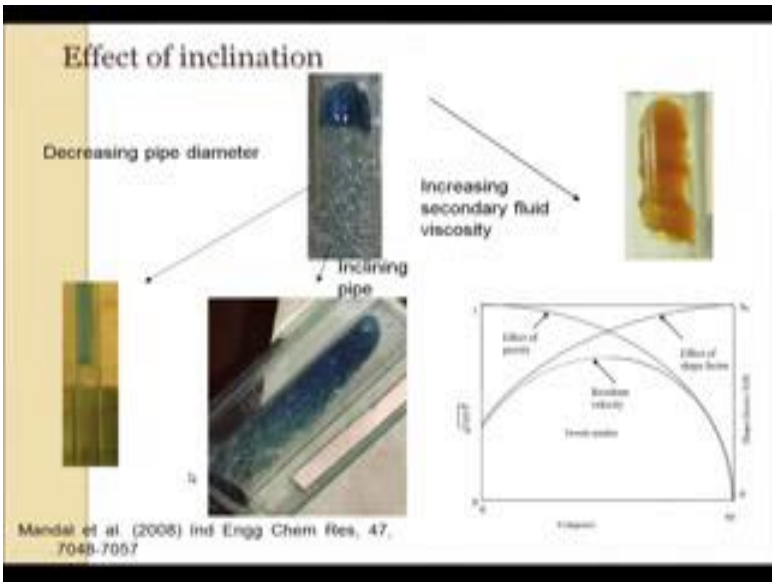

Now, we go to the next part of a discussion regarding the effect of Inclination. Now again I can show you a video in order to understand this particular influence if you observe the advance more I will start the video here, we have already seen this, now with this particular bubble if you observe the situation in an in client you what do you find. You find that the bubble in this case it has shifted to the upper portion and the liquid is draining there is a very thin liquid film of the upper portion, but the liquid is predominantly draining though the lower portion and we find that in this particular case; if you observe (Refer Time: 15:50) if you compare the 2 videos you find that the moves is slightly pointed in this particular case, therefore the raise velocity is slightly higher in this particular case.

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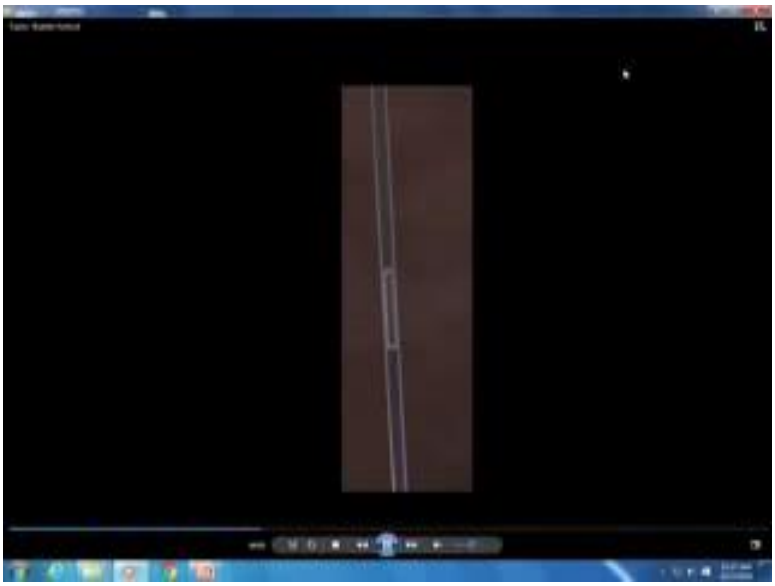

Now, this is to for macro micro every system and this is a very counter intuitive phenomena because apparently what we think is that as in a vertical tube the bubble should be raising fasters and as we incline the tube the bubble raise velocity should decrease why because as I am already mentioned, the bubble rises due to bouncy. Naturally as the buoyant effect start decreasing the bubble should also be rising at a slow slower and slower velocity, then it attains the horizontal orientation and it does not raise at all. But we find a very counter intuitive phenomena in this particular case.

We find that as we are inclining the tube definitely the gravitational component decreases, but along with that the nose of this bubble, that starts becoming more and more pointed. Since it becomes pointed it can pears through the liquid well and therefore it can rise faster. Further we find that if we compare vertical conduit and inclining conduit we find that the liquid sense which falls in this particular case is access symmetric it farms a concentric annulus around the bubble. There are greater areas of interfacial share in this case. Whereas the liquid in an inclined conduit predominantly rise sorry predominantly falls through the lower surface.

Naturally the interfacial area or area of contact with the bubble is less and as a result, it can flow much more unhinged while the while the bubble raises. Now as we all know bubble raises by downward displacement of the liquid. The faster thin liquid displaces downward the faster the bubble will raise right and this particular opposing effect of

gravity and channel shape is represented in the graph which shows that with gravity, the raise velocity should decrease or rather the gravitational effect decreases with inclination from vertical.

The shape factor shows that the bubble keeps on becoming more and more pointed till some particular angle of inclination and then of course, it again becomes rounded when we make the tube completely horizontal. Due to this opposing effect if the result in velocity is floated, we find that the velocity is not maximum and the vertical orientation rather it is maximum at some intermediate angle of inclination, which lies between say 40 to 50, 55 degrees. In this particular case the bubble raise velocity is maximum.

Well there is another interesting thing which I would like to tell you we had experimented as I have said both with gas liquid and liquid-liquid systems. Now a gas Taylor bubble raises through a liquid for both many as well as macro channels, but we find as i have already mentioned due to the increasing effect rather due to larger surface tension of the liquid, if we introduce lighter liquid into a heavier liquid, it does not raise for large channels it breaks, sorry it raises it does not raise as a single elongated bubble in larger channels. Rather it breaks in into smaller bubbles and then this bubbles the start raising. As we decrease the tube diameter the stability of Taylor bubble increases for the case of liquid Taylor bubbles rising through a heavier liquid which we shall be terming as the secondary liquid. And this also explains the cause of the existence of slug flow in reduced dimensions for liquid-liquid systems, while liquid-liquid slug flow is absent for larger channels. This is just because Taylor bubbles are not stable in larger channels and Taylor bubbles start becoming stable with miniaturisation which can be quantified by the bond number or the address number.

Generally it is said that for bond number less than 1 the liquid Taylor bubble can be found, but at the same time if you observe the re liquid Taylor bubble, which we have done in 1 of our experiments which have been performed in the multiphase laboratory of the chemical engine department, we find that the liquid Taylor bubbles they exhibit some very interesting characteristics. Firstly this experiment was done with the kerosene Taylor bubble rising though a liquid water filled vertical tube. Just like a Taylor bubbles it has a spherical nose cylindrical body and a flat teal, but we find and also we find that the water flows as a annular film fits and between the bubble and the wall. And the thickness of the film it is it gradually increases from the nose step, it keeps on increasing

in the nose region and becomes steady in the teal region. These are similar for air and kerosene Taylor bubbles.

But in this particular case what we find is, that beyond the bubble there is intense wake regions which is formed because the downward falling water comes and meets the stationary water here and as a result of the larger interfacial shear in this case. So, therefore, a large number of satellite bubbles they are sheet by the liquid Taylor bubble and this particular wake region it increases with increase in tube diameter number 1 and the other thing is that as we decrease the pipe diameter we find that the wake region disappears.

The other thing is suppose instead of a kerosene bubble we adopt a lubricating oil bubble or we the secondary liquid has a higher viscosity, In that case we find that defiantly the interface is maybe, but the wake region disappears under this particular condition. So, therefore, we find for liquid Taylor bubbles. Firstly, they are formed only for only below a tube diameter for our case of kerosene bubbles in water we find that the no stable Taylor bubbles are formed for tube diameters greater than 0.0461 meters.

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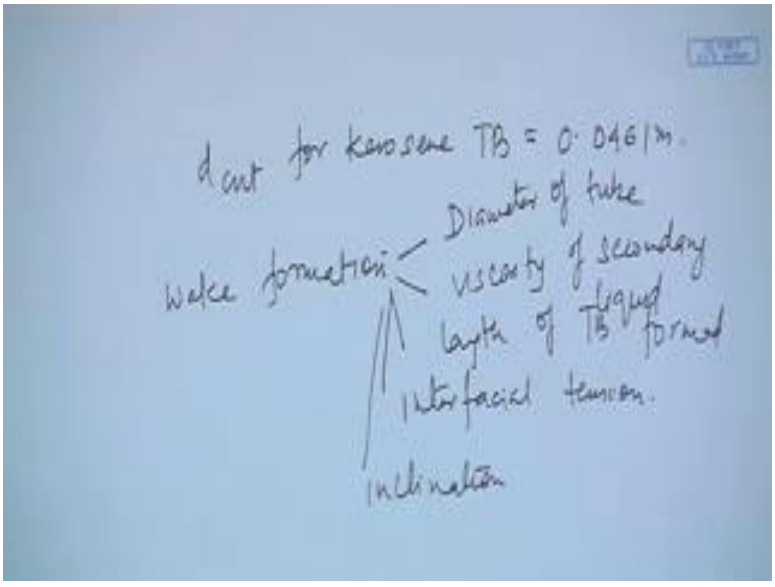

Therefore, that the d critical for kerosene Taylor bubble from our experiments was 0.0461 meters. We also found that if the experiment were performed in a perspex tube then in that case we would need a much lower rather sorry we find that the Taylor bubble does not rise in this particular case for, even for a much lower diameter or even for a

much higher diameter. For the for the case of a perspects tube we find, that that the bubble it does not rise it just ticks to the wall because we know in prospects it is comparatively more hydrophobic as compare to glass.

Therefore, the kerosene tends to stick and it does not rise for similar pipe diameters and it will only rise for a higher pipe diameter. The d critical is higher in that particular case. So, therefore, the first thing is the main thing or rather the characterising feature of liquid Taylor bubbles is number one the wake formation we know that the wake formation it increases with the length of the Taylor bubble, the larger the length the greater is the interfacial shear. More is the formation of this wake region. Secondly, is the diameter, higher the diameter greater is the formation of the wake region. Thirdly the viscosity of the secondary fluid because more is the viscosity lesser will be the wake formation and finally, the interfacial tension the higher the interfacial tension most stable is are the Taylor bubbles and as a result 2 Hepton on bubbles rising in water a less stable as compare to kerosene or benzene bubbles.

Therefore, the wake formation in a summary, it depends on 4 things, diameter of tube viscosity of secondary liquid, length of Taylor bubble formed and interfacial tension. This has been observed and here also we find it is another thing is the inclination. We find that as we incline the tube the wake formation it decreases because now in this particulars case as you observe the shear is primarily on one particular side. So, therefore, it is less the interfacial area is less so wake formation is also less, but other than that the effect of inclination is the same for an air as well as a liquid Taylor bubble. Well the same thing has been reported even for rectangular square and other symmetric conduits.

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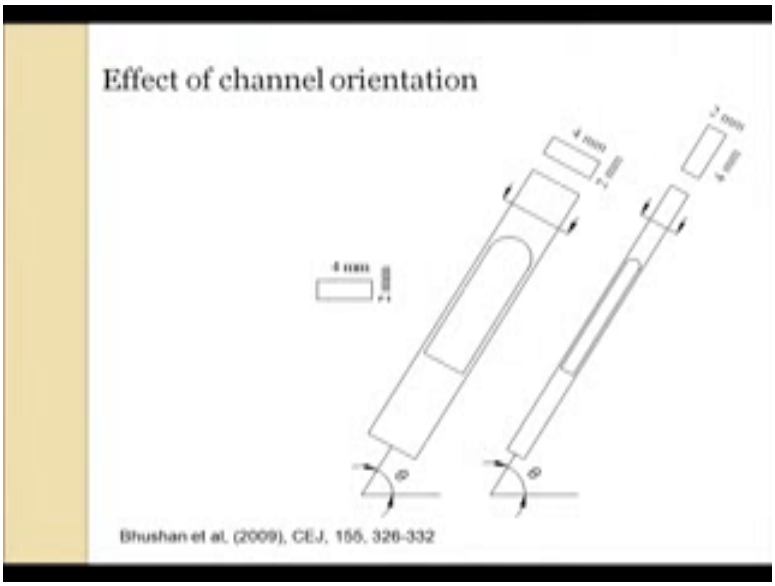

But there is one thing which I would like to discuss in the next class is a special effect of channel orientation in Rectangular Conduits.

Thank you very much. We continue with this discussion in the next class.