# **Multiphase Flows: Analytical Solutions and Stability Analysis Prof. S. Pushpavanam Department of Chemical Engineering Indian Institute of Technology – Madras**

# **Lecture – 34 Marangoni convection: generalised tangential and normal stress boundary conditions**

So, in the last lecture, we basically looked at Turing patterns and we looked at how diffusion can actually give rise to spatial instability, spatial patterns being formed provided the diffusion coefficients satisfy a certain relationship, okay and the reaction kinetics have some property of an activator species and an inhibitor species, so that was just to give you an idea about how the general theory of stability that we are doing can be actually applied to a wide variety of systems, okay.

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So, now today we will come back to fluid mechanics and we are going to look at this problem of Marangoni instability. So, what I am going to do is first talk a little bit about Marangoni instability then we will talk about the formulation of the boundary condition and then we will come and actually solve the problem of Marangoni instability okay. So, do this thing in 2 or 3 different ways, so you can get an overall picture okay.

So, this Marangoni instability, the physical system is the same as what we have seen earlier in the context of the Rayleigh Benard problem and just to refresh, the Rayleigh Benard problem was one where you had a liquid and what you are doing is; you are heating this, so this is at a higher temperature T high okay and let us say for the sake of argument that this interface, this is; this is a solid wall okay, that is a solid wall and here it is a; let us say this is gas.

So, you have a liquid layer resting on a solid wall exposed to atmosphere of gas, so there is a small difference in a sense, last time when we did the Rayleigh Benard problem both the walls were solid or both the walls were free, okay but now all I am saying is this lower fellow solid, upper fellow is free, so you have a gas liquid interface here and let us say the ambient temperature is constant at some value P ambient.

So, the first thing, which comes to mind is when I am going to be heating this, there is going to be a temperature gradient, which is established okay and beyond a certain Rayleigh number you will; which we derived last time, some 6050 for a particular boundary conditions, 1700 for some other boundary condition, you will see the onset of convection okay.

So, as the lower plate temperature increases, we see convection if the Rayleigh number is  $> a$ critical Rayleigh number and this is provided buoyancy is the one which is actually driving the convection okay, this is when buoyancy drives the convection. You remember, in the gravitational term, we had rho g and rho, we said was varying linearly with temperature and we included that.

And the first experiments actually done by Benard, they were actually with a very, very thin film okay, of liquid, so the liquid film was very thin, which means the Rayleigh number has this film thickness remember, it has the film thickness which is D and it is raised to the power cube; raised to the power 3, okay.

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So, Rayleigh number is proportional to d cube, where d is the film thickness, the point is even when Rayleigh number is of the order of 1 that is much lower than the critical numbers that we have seen, okay much lower than the critical Rayleigh number, convection has been observed. In fact, the Benard, who first observe these kind of patterns, he did not do it quantitatively, so we actually do not have any estimate of the Rayleigh number.

But the idea was that the film is very thin, so the film is very thin, we are suspecting that the Rayleigh number is much smaller okay. So, one can come up; come to 2 conclusions; one is that the theory that we did was wrong in the sense, we are predicting Rayleigh number has to be greater than 1700 for the convection okay but clearly experiments is showing that you have convection even when Rayleigh number is 1.

So, that means something is crazy okay, that means the theory is wrong and we have to go back and relook at it, maybe go back and make some more realistic models okay or the other conclusion is than the convection is actually not being driven by buoyancy but by some other mechanism, yeah, **"Professor – student conversation starts".** When the film is extremely thin and you are heating it, then the interface will deform, if convection sets in.

And if it is thin, then that will have; it will play a role in determining, we have not taken that into consideration. We did not take that into consideration in the Rayleigh Benard experiment yes, so if you remember the Rayleigh Benard experiment, well one analysis was with solid walls okay, so in that case you do not worry. So, the question is I mentioned that the

resolution of this conflict between the theoretical prediction and the experimental observation is there.

In the sense, the theoretical prediction predicts 1700 or more okay and experimentally of course, 1700 for 2 rigid walls and what we can do is; actually, do a prediction when you have one solid wall and one free surface here and I can calculate the critical Rayleigh number that will also be still high. So, the question was maybe the assumption of the interface being flat is the problem, okay.

So, if you allow the interface would deform then maybe the critical Rayleigh number would be much lower okay, so that is also a possibility, so that is one way by which you actually go about; so is it possible that by allowing the interface to deform the critical Rayleigh number will be lower that is the question okay. So, we had assumed that the interface was flat and got a high Rayleigh number critical.

What if the interface is allowed to deform? So, maybe that can result in a much lower Rayleigh number is at the point, maybe this will result in a much lower Ra critical okay. **"Professor – student conversation ends".** So, okay, the answer to this question is one has to actually do the calculation to find all the critical Rayleigh number is like, right, it is going to be still higher, it is going to be much higher on the order of 1000's, if you actually do the calculation.

But the fact that there is the another mechanism other than the density gradient comes because what people wanted; even when you have experiments being done in outer space on a space shuttle where the effect of gravity is not there, okay. People observe natural, they are not use the natural convection, they use observe convection that is even in the absence of gravity, even in the absence of this gravitational field, which is what is actually giving rise to the convection in the Rayleigh Benard problem, you can actually see convection okay.

Which means that there is another mechanism which is actually causing the convection, so for example how do you do these experiments, one is you can go to outer space and do these experiments and people have done that just to negate the effect of gravity they say look gravity is causing this problem, so I want to grow crystals; semiconductor crystals let us say in outer space, I do this over there.

And I do not have, you know any imperfection, I have very good quality crystals or you can do this experiment by just having a free fall, you just drop your experimental setup for 30,000 high in the atmosphere, I think the effect of gravity is neglected and then what you see is you do your experiment, so of course, you have very, very short window for doing the experiment but time it actually falls.

So, people have done this, the fact is that even in the absence of gravity to explain why there is another mechanism for causing this convection, you actually do seek convection, okay and that is convection occurs in the absence of gravity and it will also occur if the geometry is such that the interface is below that is you have a solid wall on top and you have the liquid film at the bottom, I mean liquid at the bottom exposed another fluid okay.

Even in that case, you do see convection patterns so these convection patterns are basically present even when the gravitational field is in the opposite direction okay, to the temperature gradient, so that means the gravitational field, the buoyancy term is not the one which is causing the convection.

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So, point is even when gravity is in the opposite direction, I mean I think that is possibly the wrong thing to write, so let me put it this way. Even when the solid wall is on top and hot and the interface is below, this means this is my solid wall and that is my liquid and this is my gas liquid interface okay and that is my gas liquid interface, we can see convection. So, you have actually hot liquid at the top; hot surface at the top and cooler surface at the bottom okay.

And that is normally a stable configuration, alright so that is we have hot wall and this is a cold interface, so here again we see convection. So, basically these are indications that there is something else which is actually causing the convection to occur okay and so the idea is what is this thing that is causing the convection to occur and that is that the Marangoni instability is basically is the other mechanism.

And in this particular convection is actually being driven by surface tension gradients okay, so what is causing this; it is surface tension gradients, what I want to emphasize here is that this atmosphere here is that a uniform temperature T ambient that does not mean that the interface here is at T ambient, there is going to be some kind of heat loss, I am not fixing the temperature of the interface, okay.

The temperature of the interface is something, which I have to find out okay and it is going to be given by a boundary condition like your Newton's law of cooling, which says that –k dt/dy equals h times T - T ambient, this is at the gas liquid interface okay and the gas liquid interface and this is the typical kind of boundary condition, we normally use, all right. The conductor flux here must be equal to the convective flux.

The point I am trying to make here is at the interface, which is you can decide, what it is?  $y =$ d, let us say, I am not fixing the temperature, I am just saying that T ambient is constant, this is infinite amount of atmosphere available, so that guy does not change. So, everywhere T ambient is constant but T can change okay. So, if temperature can vary at  $y = d$  in this direction and then what can happen is the surface tension can be is usually dependent on temperature, okay.

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So, now let us see what is going to happen because of that so, supposing the earlier problem of natural convection, we included the effect of density as a function of temperature, now we are going to include the surface station as a function of temperature okay and now we are going to see whether that can actually give rise to convection. So, I come back to this figure. This is T high that is T ambient, okay.

Now, supposing you give a small perturbation, there is a disturbance which is actually let us say; you know you have this kind of a configuration and you give a disturbance, what is going to happen is the liquid here, which is hot okay. Let us say is getting pulled up, I am not going to say that it is being induced by buoyancy, let us say there has been a disturbance which has actually caused the hot liquid at the bottom to rise up.

Now, at this point; so this guy is at high temperature, so this is at a high temperature, the interface remember is at a low temperature, so I had a particular instant of time, this guy is at a low temperature and there is a hot packet of liquid which is momentarily come up because of a disturbance. The question we are asking is how is this disturbance is going to propagate, if this guy is at a low temperature and let us say the surface tension; the surface tension decreases with an increasing temperature.

And that is the usual behaviour okay, for any liquid; the surface tension is a function of temperature and let us say, decreases with increase in temperature, which is a normal behaviour. So, the surface tension here is going to be temperature is low and the surface tension here is going to be high, so gamma is high here and the hot packet of liquid has come up here, so gamma is low at the interface the force is higher compared to here.

And therefore, there is a tendency for this liquid to be dragged along the interface because here the surface tension is less, here the surface tension is high, this guy is going to be dragged. Similarly, here I mean the hot packet has come up right, temporarily the temperature has gone up here because let us say one liquid has come up; one small packet of liquid has come here, this guy is also low and so this guy will have a tendency to come to this side.

So, basically what I am saying is the liquid will have a tendency to go towards the colder temperature because of surface tension that is high, the force there is high. Now, when that happens because of the equation of continuity because of conservation of mass, this guy has to come down okay. When one packet has gone up, this packet has to come down, so this guy will come down and this guy also has to come down eventually to fill up this space here.

So, this liquid will move, this liquid will move and you have this kind of a convection pattern. The point I am trying to make here is that I am not talking in terms of a density induced convection, I am talking in terms of surface tension induced convection okay. So, if the surface tension is not a function of temperature, then this driving force is not going to be there, this guy is not going to pull, okay.

So, therefore what I mean is; I need to include the effect of surface tension as a function of temperature in my model and it is going to occur in the boundary condition okay and then we should be able to see convection, so this is basically what Marangoni convection is all about okay. So, here surface tension decreases with the increase in the temperature, if we have hot packet of fluid rising due to a disturbance, okay.

Then, this is dragged on both sides by the cold fluid, which has a higher surface tension okay and by continuity, which is basically conservation of mass, the cold fluid comes down and we see convection. Remember, if it had been the other way, if the surface tension had actually increased with temperature, then you would not have seen this. If the surface tension at increased with temperature, this guy would actually be lower surface tension, this guy would be sorry; this guy would have been higher surface tension.

And the liquid would have been pulled from here and they were stabilized the flow, okay. So, basically what we want to do now is; talk in terms of the boundary condition, which is going to be applicable here and remember till now, we only talked about the normal stress boundary condition and basically here what is going to happen is the boundary condition that we are interested in is the tangential stress boundary condition, okay.

So, we need to basically find out how to include this boundary condition and boundary condition has to take into account the gradients in the surface tension, which is actually being caused by the gradients in the temperature. Now, the surface tension of course, is also going to be dependent upon the concentration, so for example you know that if you add a surfactant, the surface tension is going to go down, okay.

So, similar to this temperature gradient, if you also had a concentration gradient and if the concentration increases; of a surfactant increases you will have a decrease of the surface tension okay. So, the same kind of behaviour you can expect to see, so basically when you can actually add surfactants and you can, have concentration variations inducing convection okay, so that is something which people have also done.

So, in addition to temperature gradient also concentration gradients, which can actually cause Marangoni instability, so it is not just Marangoni instability is a very general thing, its talks about surface tension variations, which can be either due to temperature or concentration or anything, which can actually cause convection okay.

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So, if the motion is induced by surface tension gradients, we have Marangoni convection okay. So, what I want to do now is; this is just a background for why we have to worry about the surface tension gradient, now what I want to do is; talk about the formulation of the boundary condition at a general; let us say liquid-liquid interface. So, normally you are used to dealing with flat surfaces, continuity of shear stress okay.

So, let us look at certain things, so whenever you talk about interfaces, one thing is we say that the interface is infinitesimally thin, so there is zero thickness that means that has basically got no mass, okay. So, what that means is at the interface, we always have a force balanced that is to say the net forces were acting on the system on the interface is going to be 0 because if is nonzero that means there is going to be some kind of acceleration.

If the mass is 0, acceleration has to be infinite okay, so basically what this means is since the interface has 0 thickness, okay or negligible mass; negligible mass, negligible thickness okay, the forces acting on an element on the interface has to be 0, why? If it is non zero that means there is going to be an acceleration and which will be actually infinite because the mass is 0, okay.

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So, this okay; if not we would have let us say; infinite acceleration at the interface. So, of course, we talk in terms of a liquid-liquid interface, let us say this is liquid 1 and that is liquid 2 okay, so you have molecules all over the place and you also have molecules all over the place here. I am just to differentiate these molecules, I am using 2 different symbols, the molecules which are in the bulk, okay far away from the interface, they are going to be surrounded by molecules of the same liquid.

This guy is going to be surrounded by molecules of the same liquid, so whereas the guys at the interface, at the bottom, they are surrounded by molecules of liquid 2 and on the upper side they are going to be surrounded by molecules a liquid 1. So, what I am saying is the cause; actual cause for the forces acting on the interface is actually attributed to molecular interactions at the interface.

Whereas, here it is completely symmetric and uniform, so this guy has an atmosphere, which is only of this kind of molecule, this guy has molecules of liquid 2 partially and liquid 1, okay, so there is a difference in the environment, which the molecules at the interface is going to see and because of which; because of this intermolecular forces, you actually have this surface tension.

So, that is actually the; and if you really want to be able to predict what the surface tension is, you have to possibly go to a molecular level and come up with some kind of a theory for the description of the surface tension okay. So, point is at the interface, we have the molecules having an environment of both molecules, okay. Well in the bulk, far from the interface, molecules are surrounded by the same species, understand what I am saying.

This guy will always have molecules of liquid 2, this guy always has molecule of a liquid 1, so really experienced any net force but this guy partially liquid 1, partially liquid 2, there is a net force at the interface. So, you really want to understand what is going on, you need to go to the molecular theory and do this, okay. So, just like; we have constitutive relationships for the shear stress in terms of velocity gradient.

People have basically come up with some kind of a constitutive relationship and talk in terms of a net force like a surface tension, which is acting on the interface. Now, how valid; so how valid is this approach of just saying that there is a net force sigma, which we have to use which is acting on the interface, the validity will come by using it in developing our theory. Looking at predictions of the flow behaviour and seeing if it is consistent with experiments okay.

So, if it is consistent that means, this theory is something which you are happy with and you can use it for practical purposes, if it is not consistent, then you go back and redefine your theory and come up with introducing new properties for example. So, for example there are many people who sit down and talk in terms of no viscosity, which is a bulk property. So, there are people who talk in terms of interfacial viscosity that is there is going to be a viscosity as an interface which is actually different from viscosity of the bulk, okay.

So, depending upon the level of detail you want to get into, you will start working and including these effects, right now what we are going to do as far as our approach is concerned is; we will just say that there is a force, which is acting on the interface and this thermodynamically we want to have a consistent picture, so thermodynamically we have the argument that surface tension is basically looked upon as the work done per unit area.

So, if you have an interface with a particular area, you increase the area of the element there is more energy, which is stored. So, the thermodynamic perspective is on an energy per unit area basis but since we are more interested in the dynamics of the system, we want to use a force perspective, so we talk in terms of force per unit area, okay.

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So, basically what I am saying is the thermodynamic perspective; perspective is the energy per unit area that is what surface tension is, okay. If you increase the area of an interface, you try to have a spherical drop and you try to change the shape of the drop, the amount of energy because the sphere has the minimum area, you are going to necessarily have a larger area okay, we actually have to do work in order to change the shape of the drop.

So, that is basically your thermodynamic perspective, whereas from a mechanical perspective, we talk in terms of force per unit length okay. What I want to do is basically write down what I just said earlier that is the net forces after acting on the interface should be 0, okay. So, now let us look at an interface, which is; this is going to be tricky, so this is my curve interface and that is my normal and that is kind of tangential, that is tangential, okay.

So, this is my normal to the interface and that is the direction of tangential, so this is the view from the side, so we want to look at the thing from the top, I would have some kind of an interface like this and I am going take an area element, see I am talking about interface, so there is going to be an area element dA, okay. So, this is my area element dA and the outward normal is now outside the board, so I can show that.

But the tangents are going to be in this direction that is my tangent, so what I am going to do now is look at; let us say capital T here and T tilde, earlier I had used the T1 and T2 but now I am following Gary Leal, in his convention, so I am just going to use whatever he has done okay. So, T is the total stress tensor in the upper liquid and T tilde is a total stress tensor in the lower liquid, okay.

So, basically what this means is T is;  $-PI + tau$ , the pressure plus the shear stress coming because of the motion, T tilde is  $-p$  tilde  $+$  tau. So, on this interface, I like to write down all the forces that are acting on the system, right from this interface where the direction of the normal is given by n, okay. What is the force balance?

**(Refer Slide Time: 38:54)**

The force exerted by the upper liquid; force exerted by the upper liquid is T dot n, the total force that is all the 3 components are there, I mean if I have an interface, the direction of normal is n, what we said is T dot n tells you what the total force component is, right. This is a stress tensor, unless your outward normal is a vector, so this is going to give you a vector, so all the entire force component is there.

Now, force exerted, so this is as you are approaching the liquid from the top; the interface from the top. Similarly, force exerted by the lower liquid is going to be given by T tilde dotted n tilde but remember n tilde is - of n, okay remember, n tilde equals - n and therefore, this is - T tilde dotted with n, so this is due to the both stresses, which are acting on the top and the bottom.

In addition to that, if I look upon the surface tension force as a force per unit length along the perimeter; along the perimeter of this area element, I have a surface tension force okay and that is going to be acting along the length, along the tangential directions, so the surface tension force is given by gamma t dl, okay and this is along the perimeter, whereas this is along the area element dA.

So, these guys, this is along dA, this is along dA and now I want the net forces to be balanced okay, so I have integral, I am going to put a double integral for my area T - T tilde dotted n  $dA$  + integral over the perimeter must be = 0. So, basically this tells you all the net forces are acting on the system, I told you I want these forces to actually balance out and give me 0 okay.

I have a problem in the sense that this guy is an area integral and that guy is a line integral, so now if you go back to your Reynolds transport theorem, we have the same situation, we had a surface integral, we had a volume integral, we converted the surface integral to a volume integral by using some divergence theorem okay. So, what we are going to do now is; I like to get a boundary condition, a boundary condition which is going to be valid for every area element dA, right.

So, what I am going to do now is convert this guy into an area element and then I am going to say that this has to be valid for any dA and for every dA, it should vanish that is the argument okay. So, now for this you need to go to calculus, so I am not going to do the mathematics, I am just going to tell you what the formula is, so I am going to convert the line integral to an area integral okay.

And that means over the double integral over; that is fine and n dot del or del dot n, maybe I will write this as del dot n, yeah so this is of course, those of you are interested, we should go and make sure that you simplify the left hand side, you simplify the right hand side and show that they are equal okay, otherwise you just have to accept whatever I have said. Here, this is gradient of s or the gradient along the surface okay.

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Now, the gradient along the surface, this particular gradient s; see if the gradient is a vector for the operator, right, (()) (44:49) grad of something, I want the gradient along the surface, so what I have to do is; I have to take the complete gradient and subtract the gradient along

the normal so that is going to give me the gradient along the surface. So, the gradient of s is written as - gradient of n times n dot del.

This is the total gradient, this is the normal component of the gradient, n dot del tells you what the normal component is, multiplied by n tells you what the; this is just the magnitude of the projection with n tells you the actual component and I subtract that from the total gradient, I get my surface gradient okay. So, basically the vectorial gradient I am resolving into 2 components, which is the normal direction and the tangential direction.

And the point that is important here is that this guy; the line integral actually has 2 components; one which is along the tangential direction and one which is along the normal direction okay. So, clearly what this means is the normal stress balance will have this term contributing, the tangential stress balance will have this term contributing okay. So, what I am going to do now is substitute this back inside here and I am going to get a double integral over the area as;

So, I want this to be true for any area element, no matter which one I choose, how small I choose, which means this can vanish the integral can be 0 only if at every point, it is 0, you understand. This is true for all area elements dA therefore,  $T - T$  tilde dot at  $n +$  gradient on the surface of gamma - gamma n times del dot n equals 0 and remember this is a vectorial equation okay.

This is a vectorial equation because the gradient along the surface has got 2 vectorial components, this is a scalar but this a vector and T dot n is also a vector. So, basically I got a force balance for every element now and all I have done is the force balance converted line integral to area integral and made some arguments and I got this but when it comes actually solving a problem, I need to resolve this in the normal direction and in the tangential direction and get my boundary condition, okay.

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So, in order to find; so this is the force acting on the area element in order to find the normal component, I will dot this with n, to find the tangential component I will dot this with T and I get my relationship that I want, okay. So, to get the normal component balance, what do I do? T - T tilde dotted with n and I think I have did n dot in the earlier lecture, so I stick to that; gradient of gamma is not going to contribute; grad s is not going to contribute because that is in the tangential direction.

This is; I am doing a dot product with the n okay, so when I look at this this thing for example, look here if I do n dot grad s, I get n dot n; sorry n dot del - n dot n, n dot del, so n dot del will cancel, so n dot grad s is 0, okay, so that goes off and I have n dot n, which is again unit normal is 1, so I have - gamma del dot n equals 0 that is my normal stress boundary condition and this remember is your kind of a curvature term, del dot n.

And gamma is your surface tension, okay. If you want to actually get the tangential stress boundary condition, what should you do? Take the dot product with T and you get T dotted with n + gradient of gamma along the surface dotted with  $T = 0$ . So, what we will do is, we stop right now, we will continue from here in the next class.