Rheology and Processing of Paints, Plastic and Elastomer based Composites
Prof. Santanu Chattopadhyay
Rubber Technology Centre
Indian Institute of Technology Kharagpur

Lecture 30: Melt Fracture and other extrudate instabilities

Welcome to NPTEL online certification courses on Rheology and processing of paints, plastic and elastomer based composites. Today we are in week 5, lecture number 5.6, which talks about melt fracture and other extruded instabilities. The concept cover today will be mostly it will be centering around melt fracture and then we are going to touch upon types of melt fracture effect on I mean melt fracture I mean how melt fracture depends on laminar and turbulent flow conditions, sharkskin elastic turbulence effect of shears rates and Weisenberg number and factors affecting melt fracture, frozen in orientation drawdown, neck-in, parison sag, die lip build-up that is called drool and surface tearing. And these are all remember what I am going to talk about has some relevance or the other with some of the processing techniques that you often encounter as a problem or you have to really troubles as an in production engineer or for in the shop floor. So once again some of the keywords given here all are common I already talked about except probably angular rotation Reynolds number also I have covered.

So everything is covered if you wish to read use those keywords to search and you know web. So now coming to the point melt fracture, melt fracture is defined as the phenomena caused by excessive shear stress exerted on the molten resin as the melt fracture the name says that leads to roughness in the extruded. So melt fracture is caused by two distinct phenomena one is the pulsation in the melt pressure. So if you have that pulsation then you obviously have some roughness because of the pumping in and with the pulse and also the skin rupture happens on the surface of it.

So these two pulsation in the melt pressure are caused by basically stick slip the pressure arises some point of time it sticks to the metallic surface and then next passage next the higher stress or higher strain it tries to come out of that. So it has contact it loses contact and that happens as a periodically basically and that is what is that phenomena. In the whole if you look it at from the surface appearance you get out of melt fracture can be categorized into three types. Visually it is very distinctive if you try to go look through shark skin I mean just like a literally the name says shark skin just like a skin of a shark I mean it has a very regular undulation and very you know wavelength is longer comparatively you know and frequency is I mean less as compared comparable to the stick slip phenomena which you can see very very very very and but this stick slip appears as a patches you can see unlike this is a continuous shark skin you have it to what happened like a over a over certain periods it has certain periodicity that way I mean if you just cover the length of the extruded. Another is a gross melt fracture which is actually useless if you get this kind of a thing anyway you cannot use it.

So that is what the condition you should discard all together you cannot really do much of a troubleshoot for that. So as long as melt fracture is concerned, you may be asking yourself the question what is a shear rate responsible for melt fracture. See it is very interesting if you look it at shear stress, shear rate this is the region in between I mean there is a lower limit and upper limit of shear rate across with melt fracture happen that what I mean to say at lower shear rate region there will be no melt fracture and higher also there will be no melt fracture in between is a region of interest where I will not say interest you have to discard while processing. So this is the thing if I just try to see map it shear stress versus shear rate I can show you distinctively I mean different region where you have a smooth surface initially of course. Then you start seeing the shark skin some point of time with a pointed arrow here you can see and then at a little bit higher shear rate you can see the occurrence of stick slip and after that I mean in that particular context you see waviness.

So you can understand as you keep on increasing shear rate if you even if you can get rid of shark skin other I mean events become a little prominent and that has lot to do with the surface roughness I mean loss in surface gloss etcetera etcetera. So as an commercial point of view you do not like those kind of event because it ultimately lead to the loss of the gloss of the overall extruded basically. Now let us try to quickly recap what we learnt I mean this is a barrel and this is the die actually. So this flow in the laminar region the upper one is both the laminar region in that those regions one you can see which you expect the steady state flow and another case you can see some dead spots because of the you know sort of a vortex it forms there eddies are formed and even in laminar flow condition which we obviously want to have in the metering zone of the extruder but if you have such dead space somehow form or vortex form it can give rise to some sort of a deformatives but not the in the context of rupture which you are talking about right now. Second conditions the rupture at the engine itself if it happens.

So this can be very chaotic actually so tension stiffening melts it happens so. So similar so another thing event can also happen in the non-laminar flow condition of course these two are non-laminar flow condition it is a triggered in the die sort of a condition it happened in these regions. So it happens mostly for the tension thinning melts actually I mean with the tension if the melt thins basically and obviously in the die region there is a tension occurrence of tension because you are suddenly going from a very wider you know capillary to a lower capillary diameter. So that is what it happened so either tension stiffening there is a first one and or tension thinning both of the cases you can see some sort of a deformatives that is happening. So these are the non laminar flow originates with a very high tension stresses which accompany constant you know converging flow at the entrance or to the extrusion dies.

So this is very very important another case you have a wider gap wider capillary again it is going to a narrow capillary suddenly. So this is the possible site for non laminar flow is occurring here that you have to give it a so the forces giving rise to non laminar flow is said to originate at the empty region of the die. So this is what's happening here right over here and this triggered by the failure of adhesion at the metal interface of the die. So that is what it happens I will elaborate with the flow lines little later. Now let us try to understand and see ourselves what is Shark skin in a closer view closer proximity view.

So Sharkskin occurs at a at low extrusion rates even as I mentioned mostly the lower region is important for that it is very dependent on temperature unaffected by L by D ratio mostly and the narrow molecular weight distribution sort of a polymer has a inclination towards the Sharkskin. So always that is why it is a thumb rule that if you have a wide molecular weight distribution polymer it is it gives you a flow consistency, you can see whatever is happening in the gap will be patched up by the lower molecular weight stuff. So whatever the elastic effect happening because the high elasticity of the long chain molecule and that you know limitation will be actually patched up by the low molecular weight. So there will be overall flow continuity. So similarly there is a critical shear rate where the onset happens.

So this graph shows the shear rate of onset Sharkskin I mean the happening happen I mean there is a minimum shear rate where it happens in a polyethylene fraction as a function of die radius. So with the die radius what happens I mean your critical shear rate onset goes to a lower shear rate region basically. So that is what normally it is observed and also the critical linear velocity for a onset Sharkskin velocity means velocity of the extruded coming out. So that you if you plot it this is the line you get it with the temperature that is how it changes actually it is it increases basically. So now Sharkskin two conditions let us try to compare vice versa one is having without wall slip so you have a parabolic type of a flow.

So as a result you have a more you know extension of the molecules happening on an average. So if you consider on the contrary where wall slip happens is the parabolic forms becomes a straight type of a velocity profile more or less. So in that case average extension of the molecules in the in the slip condition wall slip condition is little less. So obviously melt fracture will be less lesser compared to that of the with slip conditions. So this is very important thing.

So you can understand if you try to see onset of you know Sharkskin happening some way or other either you do some modification in the you know polymer melt site so, that there will be a you know wall slip or else try to use some lubricants. I mean external lubricants that will come in between the polymer and the interface of the metal metallic component that is how you can circumvent it prevent it whole lot. So if the material slips very close to the die wall the velocity changes from the die wall to the extruded skin is small. So therefore the it accelerates slates that acceleration matters basically the stretching essentially elongation will flow. So as a result you will see that it is to be smoother.

So elastic turbulence is very important even if it is a laminar condition it can give rise to some sort of a you know those turbulences. So a viscous solvent let us take a polymer solution is into consideration laminar flow may be strongly modified by addition of a tiny amount of long chain molecules you see molecules and resulting in chaotic flow called the elastic turbulence. So elastic turbulence is attributed to the polymer stretching which generate elastic stress and its back reaction on the flow and the most prominent example of an unsteady flow state is called elastic turbulence and it can occur even if it is a very low Reynolds number. So that is very interesting part of it. So it is altogether is a characteristics often called elastic turbulence.

It is not the regular laminar turbulence that I talked about earlier. See now what is the effect of temperature? How does temperature you know influences the melt fracture? So if you look it at elastic turbulence here rather. So, if you look it at the shear rate versus temperature you see there is an up-turn. So critical shear rate. So you are getting some safe window more and more at higher temperature that means melt I mean elastic turbulence happening is less and less.

Reason is obvious if you increase the temperature it will be more like a fluid type rather than elastic type viscous part will dominate. But interestingly the stress part of it critical stress part of it for elastic turbulence more or less remains in different I mean with a function of temperature because stress is something to do with you have to stress the molecule. So eventually it has not lot to do with you know shear rate part of it. So, this is what it happens and also the critical stress increases steeply with decrease in molecule that is also interesting. I mean so far I was talking about that.

If you have a low molecular weight fragment obviously elastic dominance will be less and so will be the turbulence basically. So it is as simple as that. Now let us it is interesting to look it at. See this is one is the Sharkskin and this is what is a stick slip phenomena is talking about. So you remember one thing I told Sharkskin appears all throughout your extrudates basically.

Whereas your stick slip phenomena happens as a patch. So obviously from that you know periodicity as well as you know it is how frequently it is appearing you can easily discriminate whether it is because of the stick slip phenomena or it is else because of them you know Sharkskin happening because of the melt fracture. So that you will be a very well discriminated. So again apparent shear stress versus apparent shear rate. See around 100 and above apparent shear stress you get it kilo Pascal of course.

I must mention the units here. So you get to see the onset of Sharkskin and obviously the gross melt fracture which I talked about that is the worst case. It will come like a bamboo sort of a thing. So that you get it at further higher close to 1000 kilo Pascal around that region. So that is the gross difference between the you know Shark skin and gross melt fracture.

Now let us try to see practically two very common polymer plastics. One is HDPE all of you know it is a linear polymer. Another is a branched one which is LDPE. So you all know LDPE elongates more than that of HDPE. HDPE is rather steep linear sort of a molecule.

And so the elongational viscosity of LDPE is high at the first place that you can see from this plot. So HDPE is here, LDPE is this is elongational viscosity and nothing to do with the shear viscosity at this stage. So both LDPE and HDPE exhibit gross melt fracture even though they have different anti flow patterns. Both of them will show after all. With onset being at a somewhat lower critical shear stresses for LDPE than HDPE.

It is obvious. The flow curves for LDPE remains pretty much undisturbed at the onset of melt fracture while for HDPE a discontinuity appears. These are the some of the things to take away from here. The apparent shear rate increases abruptly to a certain value and then increases along a curve. At the vortex formation in LDPE is apparently related to high elongational viscosity. So that that is what it is giving you this vortex.

And as the LDPE melt approaches the entry of the capillary the stretch rate increases and so does the elongational viscosity and that is the resistance to extension which results in the large vortex formation. You see at different different shear rate this is for the HDPE you see a gross pattern which is entirely different from that of LDPE of course. So at the same shear rate you see gross melt fracture which is apparently higher for HDPE here. But in the initial stages you see there is a marked difference between these two where we expect to have a smooth extrusion. This is a PLA which is a degradable plastics.

If you look at a different shear rate the extruded you know surface at 650 pretty pretty high but 1000 is something where we actually perform extrusion and then little more 2500. So let us try to see what happens if I increase the temperature 180 to 200 you see whatever surface roughness was there that is almost vanished and if even vanishes if I try to do the extrusion at little higher temperature of course you have to keep it in mind that 220 degree centigrade PLMS start degrading. That is another part. It is not that monotonous increase of temperature will suffice the I mean solve the problem. So this this is how your extrusion profile and melt fracture varies with the increase of temperature.

This is also interesting as I mentioned it to you everything depends on the elastic to viscous response after all. So Weisenberg number is elastic forces by viscous forces. So what we are trying to do in this experiment same LDPE we try to extrude and try to have a snapshot with the you know coloring it. So that kind of a thing we do it as 1 second inverse and then 2.5 second inverse then 10 second inverse and then 80 second inverse.

So, upon doing that what we are trying to do we are increasing the Weisenberg number. So that means elastic forces are trying to dominate frequency is getting higher and higher at high frequency what happened you try to go to the from viscous to you know elastic transition I told you many times. So as a result you can see initially there is no vortex here at the corner you can see whatsoever but over shear rate the here vortex started forming even if it is only you can see 2.5 and once you go little further 10 second inverse you see it is very much there very much there. And I must mention one thing I mean those who are familiar with the fluorocarbon fluoro rubber extrusions it is very difficult to do fluoro rubber extrusion at high shear rate.

That means you cannot afford to have very fast extrusion of fluorocarbon rubber that way I mean I am talking only fluorocarbon rubber. So if you try to do it there are certain tricks and that you have to understand from the point of view what I am telling you these are the fundamentals of it. Now question is that your manager will ask you your teacher will ask you how to increase after all if you can increase the rate of extrusion the productivity increases. So obviously from commercial perspective you often will demand the high rate of extrusion velocity, average velocity across the die. So that is another challenge but understand from the point of view how to play with the Weisenberg number bottom line is that you have to reduce the Weisenberg number but you have to increase the shear rate how do you do that how do you do that.

So that is another take way I expect you ask me in the when I appear online correct. So practice affecting melt fracture there is an increase in critical shear rate for melt fracture with rise in temperature. So the product is very important tau c into mw is constant tau c is the critical shear stress above which the melt fracture happened mw is average molecular weight. So always that is one of the thumb rule you have to remember that way you can correlate one with the other. Melt fracture starts at lower shear stresses and rates and that takes place at high molecular weight regions than the low molecular weight conditions and two polymers will have similar critical points when they have similar melt velocities but different levels of branching.

So, you can understand this is also very interesting it averages out somehow and tapering the die entry that is I am talking about the angle. I will take you there when I talk about the extrusion simulating the extrusion what happens to the slip angle what happens to the you know friction of that component. So I will take you there and that tapering the die entry there will be significant improvement in the extruded quality. And this allow obtaining extremely undistorted extruded at rates much above the critical point see these are the tricks. And then however there is some internal distortion also arising out of it and the critical point may significantly increase when so called die parallel is tapered by 10 degree.

And the critical rate may also rise with the increase in the L/D ratio of die. So these are the some of the takeaways from here what I elaborated here. There is another thing frozen in orientation. See when I extruded it obviously with water or air some way or other you are trying to cool it you have a cooling fluid. So it all depends with the cooling direction flow direction of the fluid heat transfer happens more there.

So obviously that particular orientation whatever is there existing will be frozen in. So that way strength will be higher in that direction. So this is what is actually called frozen in orientation. Due to frozen in orientation plastic products display is important for plastic products not for the rubber that much because they are glossary amorphous display anisotropic that you do not want. So their property vary when measured in different direction for instance the tensile strength is more in the direction of orientation than the perpendicular direction.

And frozen in orientation also affects the impact strength. So you can understand you as a manufacturer says my impact strength is very high but what if they test it in other direction still weaker. So that kind of anomaly may happen. So you have to make sure that you have a very consistency in terms of cooling it down. So draw down is another phenomena specially it happens during blow molding.

So there is an increase in critical shear rate for a melt fracture with the rise in temperature and the product of tau c into Mw is constant as I said already. And the melt fracture starts at lower shear stresses and rates and takes place in high molecular weight regions than the low molecular weight one and rest of the things are almost pretty similar what I told. Third one is a neck-in. See the neck-in case phenomena is connected with the chill roll casting where the edge of the excluded wave has a tendency to shrink inwards. So thickness increases same thing is a calendar shrinkage it happen.

Once you calendar a sheet there will be calendar shrinkage width wise so thickness wise they will begin. So that is what in two edges here are going to be higher in thickness. So more elastic melts are less liable to neck in as they are able to maintain the tension in the extrusion direction. So, this is what it happens at some intermediate sort of molecular weight. Parison sag is interesting again it's related to the blow molding process.

So it's of its own weight it sags basically. Elastic effects can also affect parison sag which takes place during blow molding and the thinning of the parison due to its own weight when it leaves the die is known as parison sag as the name says. A portion of the sag may become may be because due to the elastic effect in chain uncoiling viscous flow also contributes the sag when in the hole, it's a kind of a creep phenomena basically. And then die lip build-up it is very common in the extrusion those who are regularly doing extrusion in the production flow you all know is nothing but although It's called drool it sounds very weird it is a kind of a deposition that it happens when it leaves the die and over time actually you are extruding over and over there will be certain depositions and that deposition varies with the flow condition flow rate basically. And only already remedies just to clean up somewhere rather remove those foreign bodies there. Surface tearing is another phenomena it happens specially for the fiber reinforced items your small fibers and in a matrix and during the flow the fibers has a open tendency to come onto the surface specially it happened if the fiber normally has a higher strength even if your matrix has a low melt strength then it try to pierce it and comes and you know accumulate and so up on the surface something like that.

So as you can see from there I can read it out surface tearing is affected by filler loading level it is most common with the fiber field extruders and the surface tearing phenomena has also been observed in the industrial extrusion and it is apparent due to the lowering of melt strength of the polymer as a filler as filler increases and the poor adhesion between the matrix and the filler and weak shear forces that allow the fiber to move at an angle that's what I mentioned. So here you can see HDPE filled with the wood pulp basically how the surfaces in the those gross manifestation of it for net HDPE smooth surface 20 percent 25 percent started having fouling the surface tearing phenomena and it becomes even worse for the as it filled more and more 50 percent 70 percent and so on. So these are the some of the things what I talked about today is very very important and this will actually help you understanding the extrusion phenomena and most importantly with this understanding it will help you for the when once you know extrude it for the troubleshooting some of the surface roughness loss in gloss ok melt fracture waviness, gross melt fracture how to handle it ok. So these are the some of the you know books and I will try to emphasize understanding rheology and technology of polymer extrusion by Vlachopoulos et al ok. So it is a very very authentic book and of course, Malkin's rheology book gives you the basic fundamentals.

So again to sum up what I talked about a melt fracture different types of melt fracture three different types so do initially and then laminar and flow conditions turbulent flow condition what are all surface defects comes. Bit on shark skin elastic turbulence effects on shear rates and I mean critical shear rates and Wiesenberg number the elastic by viscous forces if you recall and factors affecting melt fracture, frozen in orientation, draw downs particularly neck-in, Parison sag, die lip build-up which is called drool and it I think it is no more alien to you and then surface tearing effect for the fiber you know short fiber filled composite particularly it is wet compound. In the next classes we would like to talk about the plastic and rubber mixing and blending means this this slides on I mean this lecture on we will try to switch our attention to the processing side again finally, I'll try to towards the end of my lectures try to club the basic rheology what I talked about so far with the basic processing at this taking you know extrusion into as an example here. Thank you so much.