

Thermodynamics of Fluid Phase Equilibria
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Lecture – 50
Polymer Solutions

Welcome back. Today I am going to start topic on polymer solution. But let me just recap what we did last time. So, yes, in a last lecture, in the last lecture we went through the derivation of Van Laar theory where we described the derivation of basically excess Gibbs free energy.

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The image shows a digital whiteboard with handwritten mathematical derivations. At the top, it states $b_{mix} = x_1 b_1 + x_2 b_2$ with a note: \therefore no vol change upon mix. Below this, the excess Gibbs free energy is given as $g^E = \frac{x_1 x_2 b_1 b_2}{x_1 b_1 + x_2 b_2} \left(\frac{\sqrt{a_1}}{b_1} - \frac{\sqrt{a_2}}{b_2} \right)^2$. The next line shows the partial molar Gibbs free energy: $\ln \gamma_i = \frac{1}{RT} \left(\frac{\partial h_T g^E}{\partial n_i} \right)_{T, P, n_j \neq i}$. Finally, the activity coefficients are derived as $\ln \gamma_1 = \frac{A'}{\left(1 + \frac{A'}{\beta'} \frac{x_1}{x_2} \right)^2}$ and $\ln \gamma_2 = \frac{\beta'}{\left(1 + \frac{\beta'}{A'} \frac{x_2}{x_1} \right)^2}$.

And then later we showed that the activity is represented in this form ok.

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$x_1 b_1 + x_2 b_2$
 $\ln \gamma_i = \frac{1}{RT} \left(\frac{\partial \ln \phi^E}{\partial n_i} \right)_{T, P, n_j \neq i}$
 $\therefore \ln \gamma_1 = \frac{A'}{\left(1 + \frac{A'}{B'} \frac{x_1}{x_2}\right)^2}$; $\ln \gamma_2 = \frac{B'}{\left(1 + \frac{B'}{A'} \frac{x_2}{x_1}\right)^2}$
 $A' = \frac{b_1}{RT} \left(\frac{\sqrt{a_1}}{b_1} - \frac{\sqrt{a_2}}{b_2} \right)^2$; $B' = \frac{b_2}{RT} \left(\frac{\sqrt{a_1}}{b_1} - \frac{\sqrt{a_2}}{b_2} \right)^2$
 Note: $\ln \gamma_i \propto \frac{1}{T} \Rightarrow s^E = 0$

These are the forms for $\ln \gamma_1$ and γ_2 for binary liquid solution, where A dash is related to the van der Waal equation of state parameters a's and b's ok.

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non ideality of soln \uparrow with $\Delta P_c \uparrow$ $\leftarrow (P_c - P_c^0)$
 \therefore if $\Delta P_c = 0 \Rightarrow$ ideal behavior
 \rightarrow Contrary to exp
The Scatchard - Hildebrand Theory
 $s^E = 0$
 $v^E = 0$ } REGULAR SOLN
 - Essentially a modification of van Laar to remove dependency on vdw EOS

Then we talked about Scatchard-Hildebrand theory which basically depends on the same set of assumption which is basically a regular solution theory, where the s excess and v excess is going to be 0. In that case Scatchard-Hildebrand tried to avoid the the the use of van der Waal equation of state and using cohesive energy density it came up with an expression of g in this form ok.

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$$V^E = (c_{11} + c_{22} - 2c_{12}) \phi_1 \phi_2 (x_1 v_1 + x_2 v_2)$$

$$V^E = x_1 x_2 (\delta_1 - \delta_2)^2 = \gamma^E$$

$$\delta_1 = \sqrt{c_{11}} = \left(\frac{\Delta U_{\text{vap}}}{v} \right)^{1/2} = \text{solubility par.}$$

$$\delta_2 = \sqrt{c_{22}} = \left(\frac{\Delta U_{\text{vap}}}{v} \right)^{1/2} = \text{solubility par.}$$

$$c_{12} = \sqrt{c_{11} c_{22}}$$

$$RT \ln v_1 = v_1 \phi_2^2 (\delta_1 - \delta_2)^2 \quad \text{Regular soln}$$

And this they introduced volume fraction as well beside the cohesive energy density.

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$$V^E = x_1 x_2 (\delta_1 - \delta_2)^2 = \gamma^E$$

$$\delta_1 = \sqrt{c_{11}} = \left(\frac{\Delta U_{\text{vap}}}{v} \right)^{1/2} = \text{solubility par.}$$

$$\delta_2 = \sqrt{c_{22}} = \left(\frac{\Delta U_{\text{vap}}}{v} \right)^{1/2} = \text{solubility par.}$$

$$c_{12} = \sqrt{c_{11} c_{22}}$$

$$RT \ln v_1 = v_1 \phi_2^2 (\delta_1 - \delta_2)^2$$

$$RT \ln v_2 = v_2 \phi_1^2 (\delta_1 - \delta_2)^2 \quad \text{Regular soln Eqn.}$$

$$A' = \frac{v_1}{RT} (\delta_1 - \delta_2)^2$$

And in that case it turned out to be that \ln of gamma 1 is related to this where is the volume molar volume, volume fraction and the difference in solubility of the different liquids which is related to the cohesive energy density. And this regular solution equation based on this theory can be related to the van der Waal equation by considering A dash in this form and B dash in this form ok.

Now, this was what we covered now, wait we are going to just look at polymer solution ok. I will not go too much details in that, but just to give a flavour of that the theory which we looked into is all physical in nature and polymer solution later on we will be describing the Flory Huggin's theory. These based on the latest you know a concepts. However, I will not go to the derivations and rather assert the equations, but let me just describe what is a typical polymer or what is the typical properties of polymer ok.

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The slide is titled "Polymer solutions" and lists the following properties of polymers:

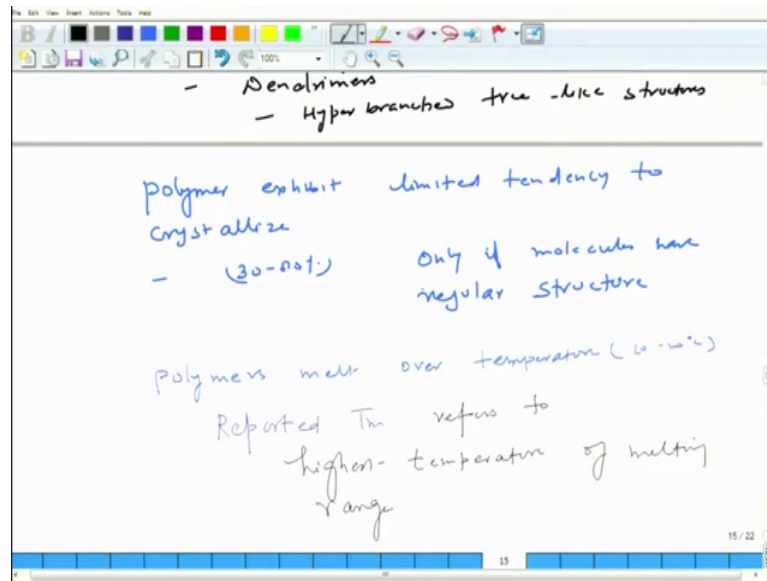
- chain molecules
 - linear
 - branched
- Homo polymers
 - all repeat units are same
- Co polymers
 - at least two or more diff repeat units
- Dendrimers
 - Hyper branched tree like structures

To the right of the text is a hand-drawn diagram of a branched polymer chain, showing a main chain of four nodes with a branch of two nodes extending from the second node. The chain ends with a dashed line, indicating it can continue.

So, if you look at polymer and compare with typical solution or typical solvent you would see that the polymers are typically made of various different chain which can be linear or which can be branched, right. So, a typical property is basically this polymers are chain molecules ok. And this can be linear, branched. Now, where the branch of react together the cross linking occurs in the polymer ok. Then you can have a homo polymer, where all repeat units are same they have copolymer where you have at least two or more different repeat unit ok. Then you have this dendrimers which are hyper branch tree like structure.

Now, the size of the polymer plays a significant role in the properties of the solution, and essentially typical properties such as or typical forces such as dispersion and (Refer Time: 04:50) bonding in such case plays a significant role and it basically governs the thermo physical property of the polymeric solutions. So, the size plays a big role ok.

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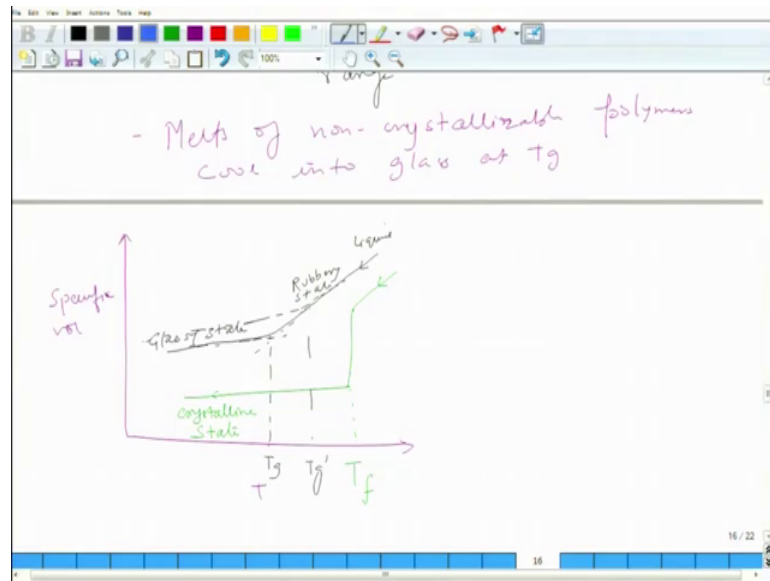


Now, these polymers can exhibit different kind of structures also. So, polymer exhibit difference structure, but it has limited tendency to crystallize. Now, usually 32 percent for 80 percent a limited crystallinity is possible, only if the molecules have regular structure ok.

So, usually that means, that most of the time the polymers are amorphous in nature ok. Now, usually the polymer melt or a temperature range, it is not like it does not it has you know it is 10 tensely melt at a specific temperature usually it has a some range of the temperature. So, polymer melts over temperature ok. So, the range could be 10 to 20 degree Celsius and the reported value of T_m , the usual reported value of T_m refers to highest temperature of the melting range ok.

The other important thing is that the melts of non crystalline or crystallisable polymer, if you cool it then it can take a glass structure at glass temperature which is called T_g ok.

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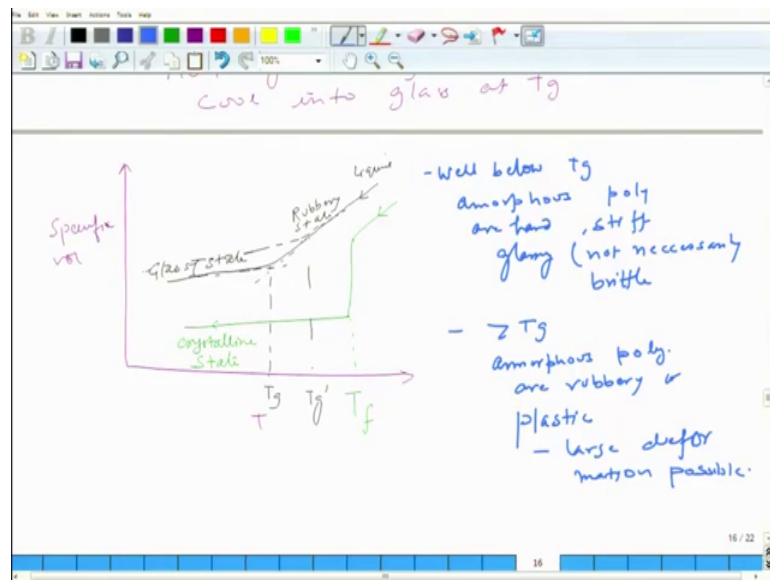


So, let me just draw typical specific volume or specific volume versus temperature and let us assume this case a typical way if it consider usual solution than this may be the case where the specific volume as you decrease the temperature and it drops at typical freezing temperature attains usually the crystalline straight, ok. So, that is a, that is a usual liquid, but in case of the polymer you would see a little different scenario. So, it may have same system or behaviour like this ok.

So, this is a let us say liquid and you are quenching it and reducing the temperature and at this point it can shave change the slope ok, and this change in the slope basically indicating of glass temperature and this is basically the thing, but glass glassy state ok. Now, it depends on the rate at which you are quenching it.

So, for example, if you quench in a different way it may happen that it occurs somewhere here so that means, you can have different different glass temperature depending on the quench right. Now, typically this is the liquid and at this point it will be some something like a rubbery state ok. So, let me just give the salient features of this different state.

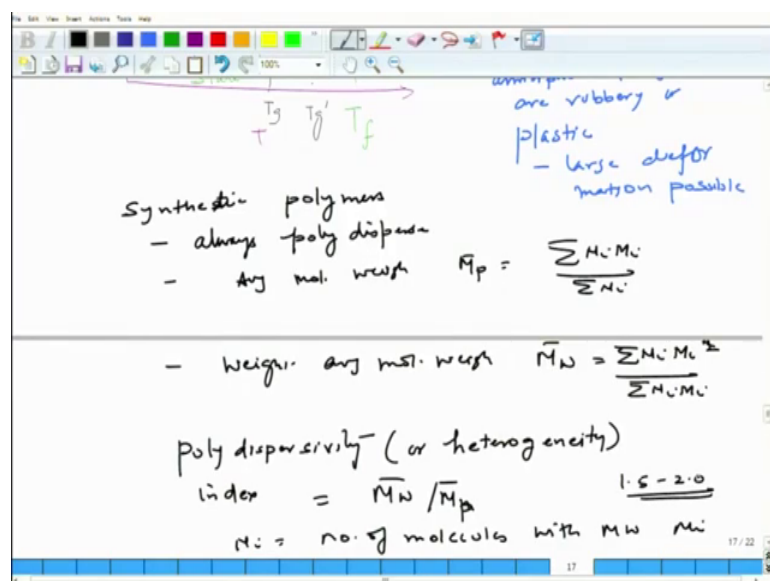
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So, well below so, below T_g a amorphous polymer usually as a hard, stiff and glassy ok. So, it need not be not necessarily brittle ok; Now, well above T_g the amorphous polymer polymer they are rubbery or plastic ok; that means, there is large deformation is possible ok.

Now, if you consider synthetic polymer, synthetic polymers are usually poly disperse, so they are poly disperse ok.

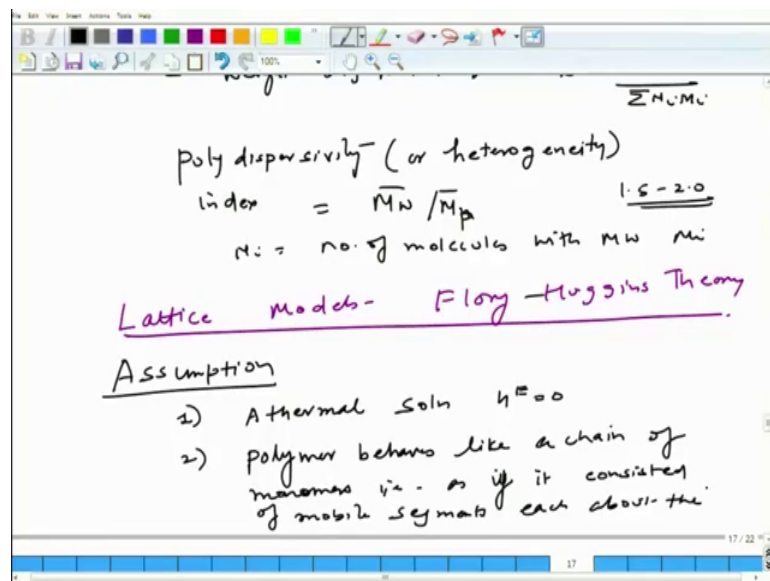
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And then the how do you measure this poly dispersity, dispersivity by considering average molecular weight. Let us say M_p which is going to be summation $N_i M_i$ divided by summation N_i , where N_i is the number of molecules with molecular weight M_i or you can consider weight average, weight average molecular weight which is going to be something like this M_i^2 divided by summation $N_i M_i$ ok.

So, and poly dispersivity sometimes it is also called heterogeneity index is given by M_w average, M_n average ok, so M_p average. So, here N_i is again this is the number of molecules with molecular weight M_i , ok. Now, this poly dispersivity index typically is in the range of 1.5 to 2.0 ok. So, that is about a brief description of the polymer polymeric system. So, what about the the theory for polymeric solutions, a popular theory is basically given by Flory Huggin's theory which is based on the lattice models ok.

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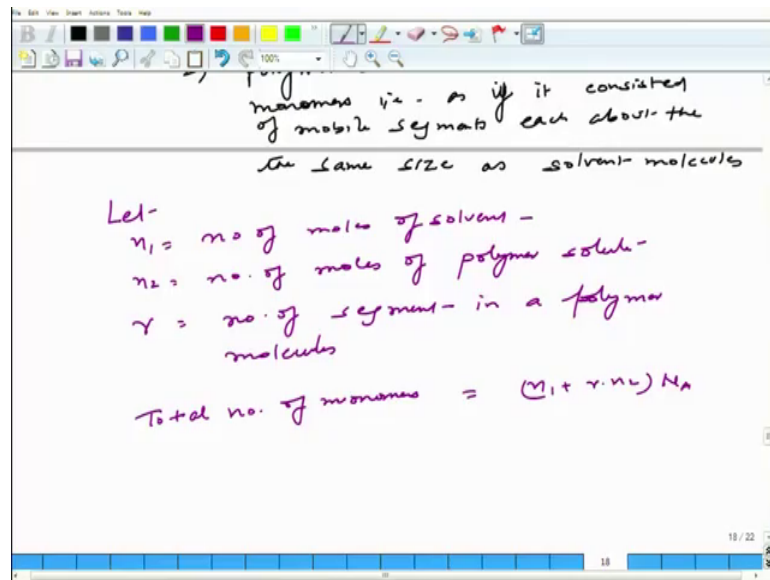


So, what are the assumptions for this? So, the Flory and Huggin's they have taken the assumption as follows. The first important assumption is basically that the system is a thermal so that means, solution is a thermal. So, essentially that its deviating from the typical of my discussion we had last time where we Van Laar or Scatchard-Hildebrand solutions, that is regular solution theory.

We have basically the excess entropies also considered to be 0, but here in this case for Flory Huggin's the solution is only a thermal solution. And other important assumption is

that the polymer behaves like a chain of monomers that is as if it consisted of mobile segment each about the same size as that of the solvent, as solvent molecules ok, so which essentially means that whatever the size of the solvent molecule the monomers or the polymer have the same size. So that becomes easy for doing all the analysis ok.

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So, let us consider n_1 has number of moles of solvent, n_2 is number of moles of polymer solute and that is say r is number of segment in a polymer molecules ok.

So, given this what is the number of monomers? That would be n_1 plus r times n_2 , and this is number of moles. So, we multiply by N_A have a good rule number. So, that would be the case of total number of monomers.

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The image shows a whiteboard with handwritten notes. At the top, it says "Total no. of monomers = $(n_1 + r \cdot n_2) N_A$ ". Below that, it says "Define vol. fractions" and gives two equations: $\Phi_1^* = \frac{n_1}{n_1 + r n_2}$ and $\Phi_2^* = \frac{r n_2}{n_1 + r n_2}$. A horizontal line separates this from the next section, which says "Flory - Huggins theory says" and gives the equation $\frac{S^E}{R} = -x_1 \ln\left(\frac{\Phi_1^*}{x_1}\right) - x_2 \ln\left(\frac{\Phi_2^*}{x_2}\right)$. The whiteboard has a toolbar at the top and a slide indicator at the bottom right showing "19 / 22".

Now, for simplicity we also defined volume fraction. So, that would be n_1 plus n_1 , $r n_2$ putting starred and this is $\Phi_2^* = \frac{r n_2}{n_1 + r n_2}$ ok. So, this is the definition of volume fraction ok.

Note that each monomer has same size and that is why it should be straightforward ok. So, now, what does follow Flory Huggin's theory says?. So, he says that S^E by R minus is basically nothing but minus of $x_1 \ln\left(\frac{\Phi_1^*}{x_1}\right) - x_2 \ln\left(\frac{\Phi_2^*}{x_2}\right)$ and then Φ_2^* divided by x_2 . So, that is what Flory Huggin's theory says as far as the intro excess entropy is considered.

And this you can derive it using the lattice model considering that the polymer occupies lattice space, lattice points of space, but we will not go through the derivation of this ok. We just asserting this expression ok. Now, you can also rewrite in terms of mole fraction, you can also rewrite this expression replacing x_1 in terms of R ok.

So, since x_1 is n_1 times you know you can also consider x_1 in terms of this moles and you can simplify this expression and bring the R which is the number of segments in the polymer solute.

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$$\frac{S^E}{R} = -x_1 \ln \left(\frac{\phi_1^*}{x_1} \right) - x_2 \ln \left(\frac{\phi_2}{x_2} \right)$$

$$= -x_1 \ln \left[1 - \phi_2^* \left(1 - \frac{1}{r} \right) \right] - x_2 \ln \left[r - \phi_1^* (r-1) \right]$$

$$\frac{g^E}{RT} = \frac{h^E}{RT} - \frac{S^E}{R} = - \frac{S^E}{R}$$

$$= x_1 \ln \left[1 - \phi_2^* \left(1 - \frac{1}{r} \right) \right] + x_2 \ln \left[r - \phi_1^* (r-1) \right]$$

$r > 1 ; \quad g^E/RT < 0 \Rightarrow$ negative deviation from Raoult's law

So, if you do that without showing the detailed analysis I am just writing the expression here minus $x_2 \ln r$ minus ϕ_1 , r minus 1 ok. So, this is the expression. Now, we know that this is a thermal solution as far as the Flory Huggin's theory is considered. So, g^E by RT is h^E by RT minus S^E by R which is nothing, but S^E by R . So, now, you have the handle to g^E . So, this is same as this ok.

So, given this that you have this g^E in terms of minus this would be your this would be your nothing, but $x_1 \ln \left[1 - \phi_2^* \left(1 - \frac{1}{r} \right) \right] + x_2 \ln \left[r - \phi_1^* (r-1) \right]$ ok. Now, for r greater than 1 ok, you can do the analysis g^E by RT is less than sorry less than 0 this is imply this is a negative deviation from the Rault's law ok. So, this is different from what basically the regular solution theory talks about ok. For the case of non thermal or basically thermal solution where basically your h^E is not equal to 0.

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for $\gamma > 1$; $g^E/RT < 0 \Rightarrow$ negative deviation from Raoult's law

For non-athermal solution, customary to add a van Laar/Scatchard-Hildebrand term

$$\frac{g^E}{RT} = x_1 \ln \frac{\phi_1^x}{x_1} + x_2 \ln \frac{\phi_2^x}{x_2} + \chi \phi_1^x \phi_2^x (x_1 + x_2)$$

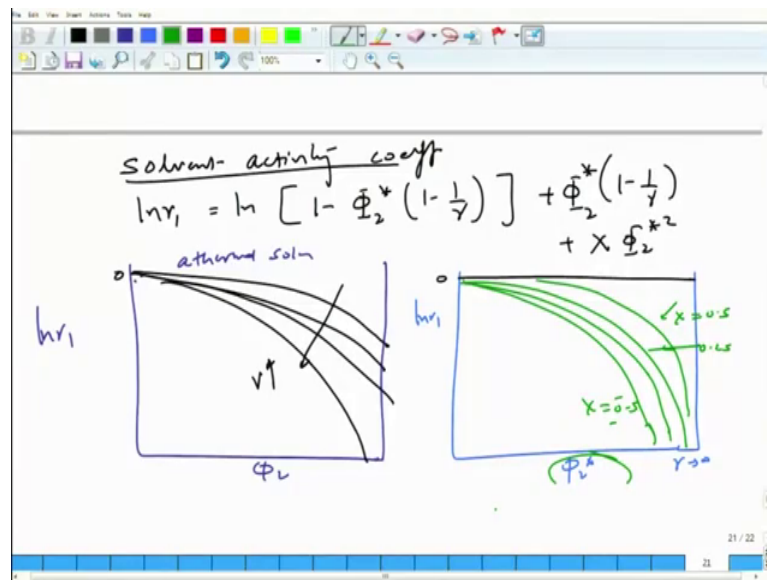
$\chi \rightarrow$ Flory interaction par. $\approx \omega / (K T)$
 - determined by intermolecular forces
 exchange energy between solvent mol. & polymer segments

You can add a van der Waal or Scatchard-Hildebrand term to g^E ok. So, let me just write down here for non a thermal solution it is basically customary to add Van Laar or Scatchard Hildebrand term. So, in that case your g^E or RT is ok. So, I can use the earlier term here which was this right. So, g^E/RT is $x_1 \ln \frac{\phi_1^x}{x_1} + x_2 \ln \frac{\phi_2^x}{x_2} + \chi \phi_1^x \phi_2^x (x_1 + x_2)$ right. So, I am going to use that.

And now, since it is a thermal. So, this is basically nothing, but your minus of s^E by r . Now, for a thermal I am going to add this term ok. So, this is due to the Van Laar Scatchard-Hildebrand term ok. So, in this case this χ parameter is called Flory interaction parameter, and it is determined by the intermolecular forces. So, this is related to the intermolecular forces ok, approximately ω by $K T$ determined where ω is basically nothing, but exchange energy between solvent molecule and the polymer and polymer segment ok.

So, now, given that you have this g^E of x available you can obtain the activity as we have done in the earlier case. So, of course, I have not gone through the derivation in details, but something which you can directly use it in order to solve the polymer solution problems.

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So, I am going to write down the solvent activity coefficient in this case activity coefficient is given by $\ln \gamma_1 = \ln \left[1 - \phi_2^* \left(1 - \frac{1}{r} \right) \right] + \phi_2^* \left(1 - \frac{1}{r} \right) + \chi \phi_2^{*2}$. So, this is a solvent activity coefficient.

Now, if you would like to see how this is going to be affected by the segment length, we can actually directly use this for a given some chi parameter. So, this is typically the behaviour of $\ln \gamma_1$ as a function of ϕ_2 for different r values. So, this is for the case of a thermal when we consider chi to be 0, ok. So, when you consider a thermal solution you will see something like this ok, where r increases along with that ok, for the case of this would be corresponding to our infinity.

Now, you can consider another case where you have again $\ln \gamma_1$ with ϕ_2 ok, where r is infinity in the sense that is infinitely long chain and then we are going to consider different chi. So, this is around this was around 0, this is around 0 and we are going to get something like this. So, it is not perfectly you know in scale, but this is something where you know the guy in this case is 0.5 and this is reducing here this is somewhere is 0 and then chi can be negative also in that case for example, chi could be in this case 0.5, ok.

So, you see it is not changing much as far as the overall nature is concerned ok, but it is relevant that to understand that it has a significant effect on their activity values. Particularly it also depends on this if you see here the volume fraction ok, of the polymer

polymeric solute ok, in this case. But again the point of description of this thing was to just to illustrate the change is it will have typically and the more details you can have you can get into some book the textbook has more detail information on it, and the idea was not to get too much in details within this course, but to illustrate what you would like to do if you have a polymeric solution and how to solve for simple problems.

So, if you if you are interested to get into more of this you should look at chapter 8 of the test book where a figure let us say 8.6, 8.7 illustrate the use of Flory Huggin's theories to explain activity coefficients of heptanes and; heptane and polyethylene solution and similarly for other cases ok.

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$$\ln \gamma_1 = \ln [1 - \phi_2^* (1 - \frac{1}{r_2})] + \phi_2^* (1 - \frac{1}{r_2})$$

$$\ln \gamma_2 = \ln (1 - \phi_2^*) + \phi_2^* + \chi \phi_2^{*2} + \chi \phi_2^{*2}$$

$$\Rightarrow \ln \gamma_1 - \ln \phi_1^* - \phi_2^* = \chi \phi_2^{*2}$$

Now, I will consider a specific case now. So, let us consider our infinity in that case log of gamma 1 the activity of this solvent can be reduced, ok. So, in this case r is infinity then you have ln gamma 1 is ln 1 minus phi 2 star plus phi 2 plus chi phi 2 star, ok. Now, this further can be simplified ok, and I can write this as ln phi 1 plus phi 2 plus chi phi 2 square, ok.

So, this would mean that ln gamma 1 minus and then phi 1 star minus phi 2 star is chi phi 2 star ok. And now, I can divide this phi 2 here, this means that ln gamma 1 minus ln phi 1 star minus phi 2 star divided by phi 2 is nothing, but chi phi 2 star.

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$$\begin{aligned} \gamma \rightarrow \infty \quad \ln \gamma_1 &= \ln [1 - \phi_2^* (1 - \frac{1}{\phi_1^*})] + \phi_2^* (1 - \frac{1}{\phi_1^*}) + \chi \phi_2^{*2} \\ \ln \gamma_1 &= \ln (1 - \phi_2^*) + \phi_2^* + \chi \phi_2^{*2} \\ &= \ln \phi_1^* + \phi_2^* + \chi \phi_2^{*2} \\ \Rightarrow \ln \gamma_1 - \ln \phi_1^* - \phi_2^* &= \chi \phi_2^{*2} \\ \Rightarrow \frac{\ln \gamma_1 - \ln \phi_1^* - \phi_2^*}{\phi_2^{*2}} &= \chi \end{aligned}$$

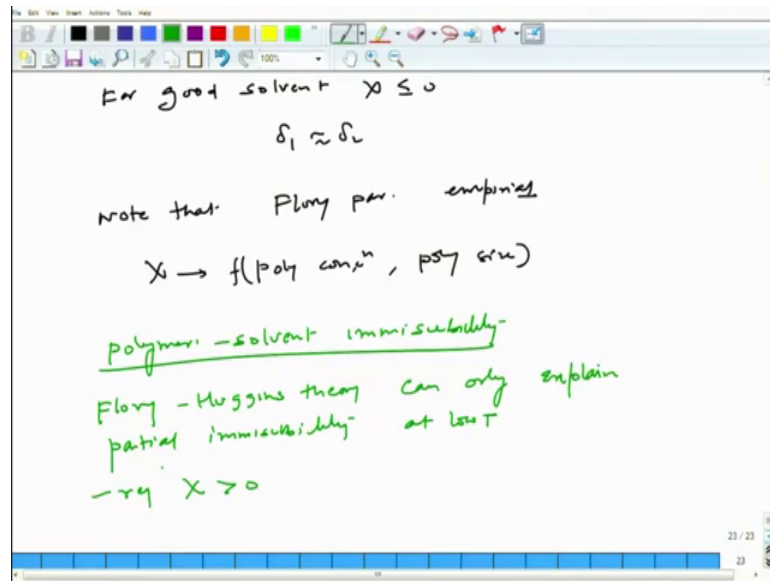
Compare with Scatchard-Hildebrand theory, we find

$$\chi = \frac{v_1}{RT} (\delta_1 - \delta_2)^2$$

Now, this is something which we can compare with the Scatchard-Hildebrand. So, if you compare with Scatchard-Hildebrand theory then you can find that that chi, so if you arrange it and then you compare with the Scatchard-Hildebrand theory we find that chi is nothing, but v_1 by RT the difference in solubility to the square ok.

Where we know this v_1 is nothing, but the molar volume of the solvent 1, and delta 1, delta 2 as a solubility parameter of the solvent and polymer respectively. So, this is something which is a beautiful way to obtain the chi parameter of Flory interaction parameter using the molar volume and solubility parameter of the solvent and polymer solute.

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So, for good solvent usually the chi parameter is less than equal to 0. Now, since chi using this is related to this which essentially means for a good solvent your solubility of solvent and the polymer should be all the same order ok.

Now, this parameter which we have obtained here or written here is rather empirical ok. So, note that Flory parameter empirical ok, and if you try to find out this chi from a polymer concentration and the polymer size dependent or if you try to find out the chi based on the neutron scattering and from calamity it will find to be different in. So, usually these are not consistent, and chi essentially is dependent on the polymer solution a polymer concentration and is also polymer size dependent. So, chi is a usually dependent on some polymeric concentration and polymer size ok.

So, in spite of that there is a limitation as far as the Flory Huggin's theory is concerned there is a large usage of such a theory for solution polymeric solution and thus is very popular ok. Now, if you are interested to find out immiscibility. So, if you are interested in polymer solvent immiscibility ok, then the polymer or the using this Flory Huggin's theory you can only obtain partial immiscibility.

So, Flory Huggin's theory can only explain partial immiscibility of polymer solution system at low T, ok. So, it requires chi to be greater than 0. So, you can do the same analysis as we have done for earlier to find out the immiscibility you look at the second derivative of g of delta g mix.

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$$\frac{\partial^2 \Delta_j^{mix}}{\partial x_1^2} < 0 \Rightarrow \chi > \frac{1}{2} \left(1 + \frac{1}{\sqrt{r}}\right)^2$$
 Incipient instability occurs at

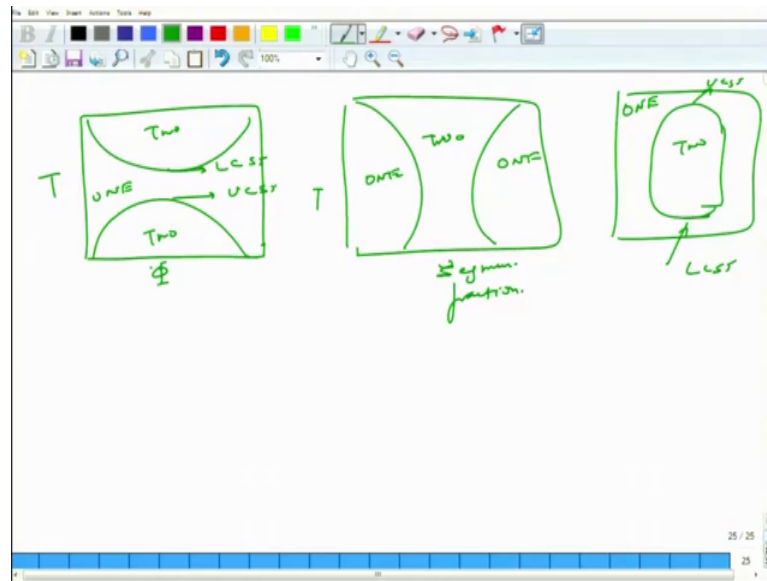
$$\chi = \frac{1}{2} \left(1 + \frac{1}{\sqrt{r}}\right)^2$$
 Corresponding critical composition $\phi_c^{*c} = \frac{1}{1 + \sqrt{r}}$
 $r=2 \rightarrow$ analysis for Margules eq
 $\chi \propto \frac{1}{T}$ \rightarrow predicts UCST

So, let us do that. So, if you find out this should be less than 0 which essentially means chi should be greater than or equal to $\frac{1}{2} \left(1 + \frac{1}{\sqrt{r}}\right)^2$. So, incipient instability; that means, when it is equal occurs that chi is equal to $\frac{1}{2} \left(1 + \frac{1}{\sqrt{r}}\right)^2$, so this should be plus plus 1 by square.

So, in that case the corresponding critical solution also a critical composition corresponding critical composition ϕ_c^{*c} it is $\frac{1}{1 + \sqrt{r}}$, ok. So, this comes directly from this analysis. So, let me make just last few comments. If you put r equal to 1, if you note here if you put r equal to 1 this will give us the same analysis as done for Margules equation.

Now, recall that chi parameter which we have shown here is proportional to $\frac{1}{T}$. So, in that case the Flory Huggin's theory is associated with only the upper critical solution theory ok. So, it only predicts the upper critical solution theory, but in reality. So, it predicts only upper critical solution, so sorry upper critical solution temperature. So, Flory Huggin's theory is associated with an upper critical solution temperature; that means, it predicts only the one such case.

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But if in reality you might see different kind of critical solution temperature. So, for example, you can have this where this is LCST, this is UCST, this is two phase, this is two phase and this is one phase and this is a function of T versus solvent segment fraction of polymer. The other case where you can have something like these this is two phase, this is one phase, and you can have something like this also, closed this LCST, this is UCST, two phase and this is one phase ok. Again this is T versus solvent or segment fraction ok.

Now, these are the keys which is for binary polymer solution and this is something which tells you that for such a scenario the Flory Huggin's theory does not work very well because Flory Huggin's associated with only upper critical solution temperature. Now, so this was kind of a brief introduction of the polymer solution that, particularly we introduced the Flory Huggin's theory which was based on which is based on lattice model.

And this will be the end of the liquid solution or theories for those solutions. What I am going to do in the next lecture is basically take up few examples to illustrate how to solve such a problem using the activity models.

So, I will see you in the next lecture and that will be the last lecture of this particular course. So, see you there.