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# Module No. # 8 Lecture No. # 20 Particle Cohesion: Forces

Welcome to the twentieth lecture in our particle characterization course. In the previous lectures, we have been focusing on particles that are suspended in a medium and the types of interactions that these particles have with the surrounding medium. In particular, we have been looking at aspects of particle to surface adhesion and also removal of particles from surfaces.

Now, in this lecture, we are going to focus on inter particle adhesion, that is, adhesion between particles that are suspended in a fluid. Another name for this is cohesion. The cohesive behavior of particles that are suspended in a fluid is very crucial in determining, particularly the flowability characteristics of the suspension.

In many industries, we have situations where particles are suspended either in a liquid or in a gas, and they have to be made to flow in a certain direction, at a certain velocity, at a certain flow rate, towards receptacle for another surface and so on.

So, it is, it is very important for us to understand how this system is going to behave, that is, the combination of the fluid and the particles that are suspended in a fluid, and it turns out that the degree to which particles that are adjacent to each other, try to interact with each other, determines the flow characteristics of the suspension itself, and so, we need to understand their reaction mechanisms as well as the interaction forces between adjacent particles, in order to characterize how the suspension will flow.

Now, when we look at inter particle forces of attraction and repulsion, many of them are common to what we had discussed earlier in the context of particle to surface interactions. For example, Van der Waals forces that we discussed in detail early on, in the context of particle to surface interactions, are obviously also important in the context of particle to particle interactions.

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The Van der Waals forces in the case of a particle to particle interaction can be written again in terms of Hamaker constant A 132. If you recall our definition of 1 2 and 3 is one will be, in this case, particle 1; 2 will be particle two; and three is the intervening medium - liquid or gas.

So, Hamaker constant multiplied by the distance of separation between them also enters your equation as 12 times. If the distance is some S 0, it is divided by twelve times S 0, and the diameter of the particles essentially representing them as equal sized particles, will again enter in the numerator as a sized term. So, just like the Van der Waals force of interaction between a particle and a surface, the force of interaction between two adjacent particles also shows a linear dependence on the particle size.

Now, this is for the case where the two particles are of equal size, particle size d p. When you have two different diameters - dp 1 and dp 2, then you essentially define and equivalent diameter, a mean diameter. So, if you have two particles that are of sizes dp 1 and dp 2, you can define an effective diameter 1 over dp bar, which is equal to 1 over dp 1 plus 1 over dp 2 and that will serve as the effective diameter that governs the Van der Waals forces of interaction between these two particles.

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So, if you look at the Van der Waals forces molecular interaction, the mechanisms and the forces are really not very different from the forces of interaction between a particle and a surface. Similarly, if you look at electro static forces, and again, let us say that you have two particles of charges, - q 1 and q 1 - then the electro static force of interaction between them will be q 1 q 2 by four pi epsilon r epsilon 0 times s squared - where q 1 and q 2 are the charges on the two particles; epsilon r is the dielectric constant of the medium; epsilon 0 is the permittivity of free space and s is the distance of separation between the centers of the two particles. So, again, electrostatic forces between a particle are very very similar in their behavior to a electrostatic forces between a particle and a surface.

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So, what is the unique about particle to particle interaction? Why do we need to study that differently from particle to surface interactions? The primary reason for this is - this cohesive behavior. What do we mean by cohesive behavior? When you have a suspension, where you have a fluid and you have various particles that are suspended in the fluid. There are various modes of interaction between the adjacent particles. In the simplest case, they do not even know that each other is there.

So, the behavior of every particle can be taken to be independent of the behavior of any other particle in the system, but in reality, there is always a tendency for two particles that are near each other to try and find each other, and when that happens, you first start forming a bridge between two adjacent particles, it is called a liquid bridge.

And when that happens, when you have bridging between two particles, this is called the pendular state of cohesion. What will happen next? A third particle will be brought into this system. So, when you have three particles that are bridged by the surrounding fluid, that is called the funicular state of cohesion, and finally, the next state would be one that involves, let us say four particles.

When that happens, that is what we call the capillary state of cohesion. Now, when this bridging extends beyond four particles and starts encompassing a larger number, so, more than four particles now begin to be bridged by the surrounding fluid. That is called the droplet state of cohesion and it turns out that the cohesive behavior between the particles very much depends on what state of cohesion they are in. For example, when you have no cohesion as I said that is a simplest case, so each particle can be treated as a hart sphere that is moving on its own in the surrounding fluid.

When you have two particles that are bridging, - pair wise bridging - the pendular state, that is the state of minimum cohesion in the fluid. So, there is cohesive behavior but it is reasonably mild. As you go into the funicular and capillary phases, the extent of cohesion begins to increase, and therefore, you have to start treating the particle phase virtually as a continuous phase that is present within another continuous phase.

In other words, you have to start following the two phase model in order to characterize the behavior of such suspensions, and by that time, you get to the droplet state, where there is larger than four particles that are essentially bonded together, it becomes highly cohesive behavior and you cannot describe the motion of such a suspension without understanding how this droplet behaves as a separate phase that is contained within the microscopic fluid phase. So, that is what makes cohesion interesting. These are behaviors that you do not normally encounter in the case of particle to surface interactions.

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Now, in terms of forces of interaction, - cohesive forces - we already talked about Van der Waals forces and electro static forces. In addition, you have what are known as capillary forces. Now, the capillary force itself has two components to it - one is surface tension related and the other is pressure differential related. In addition to that, you have viscous forces, and here, again you can sub classify them as normal and tangential, and finally, you have spring like forces and this also you can classify as normal and tangential. So, we will look at what these are in more detail.

But the point is that in order to describe cohesive bonding between two particles that are in solution, you have to characterize all of these forces and the net force that is acting will be a sum of all these forces that are acting on particle that are suspended in a solution. So, let us look at forces three, four and five in a little more detail.



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So, what do we mean by capillary forces? Let us take the situation where you have particle 1 and particle 2, and there is a liquid or fluid bridge between them. Now, the angle to the centroid of the particles that the meniscus makes this angle is called beta or we still on, and, the, as the angle beta becomes smaller and smaller as beta tends to 0, the force essentially, basically, this, this meniscus will become thinner and thinner. So, as you can imagine, there will be less and cohesive bonding between the forces. So, as beta tends to 0 F capillary will tend to 0, and as beta tends to ninety degrees, F capillary will tend to its maximum value.

So, the ninety degree corresponds to the case where essentially the liquid bridging extends all the way to the outer perimeter of the particles. So, beta is now ninety degrees. So, this represents the case where we have complete betting. So, this is a case where you are in to the so called capillary or droplet regime of cohesion.

So, whether you are in the pendular of funicular or capillary or droplet state, depends a lot on this parameter beta, which is the also called the half fill angle or the angle of cohesion between adjacent particles. So, when we look at the forces now between these particles, there are really two components to it - one is just due to the surface tension, that is associated with the fluid; that is bridging the particles, and the other is associated with the pressure differential delta p across the fluid to air interface, that is surrounds this suspension.

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So, you have to split the capillary force into its two components - F surface tension plus F pressure differential and you write those as 2 pi r p gamma times sine square beta plus pi r p square delta p sine square beta at 2 pi r p gamma sine square beta represents surface tension force and pi r p square delta p represents the circumferential force multiplied which is the pressure differential multiplied by the circumferential area of the particle or you can simplify this and write it as 2 pi r p gamma sine square beta times 1 plus r p delta p over 2 gamma.

Now, this parameter r p delta p over 2 gamma is represented as h star and it is stands for what is known as the dimensionless mean curvature, which is a parameter that can be extracted from the basic Laplace young equation governing the elastic behavior of these particles in suspension.

So, the expression for the capillary force itself can be written as 2 pi r p gamma sine square beta times one plus h star and it can be evaluated for essentially any pair wise combination of particles that are suspended in a fluid.

Now, the viscous forces again can be split into a normal force and a tangential force. The F viscous normal, the normal contribution of the tangential force is given by 6 pi mu r p v normal times r p over s - where r p against the radius of the particle; s is the distance of separation between the particles; v N is a relative normal velocity between two particles that are in suspension.

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So, if you have a particle, two particles, their velocity itself can be de convoluted into a tangential velocity and a normal velocity, and the delta between the normal velocities of the two particles enters this equation as v N, and similarly, the delta between the tangential velocities enters the equation for the tangential viscous force which is given by 6 by mu r p times V t times 1 plus point 0.5 logarithm of r p over s.

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So, in all of these equations, one of the things, we, you will notice is that if you look at the equation we have written so far, most of the adhesive forces still have the linear dependence on diameter, just like we described earlier when you have particle to surface adhesion forces, where the adhesion force essentially scaled as the size of the particle, you will see, that if you look at the cohesive forces as well, the surface tension force clearly has an r p dependence.

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However, the pressure differential, because it is an area effect has a r p squared dependence. So, the net of this term plus this term will give you a dependence of capillary force on particle size that is somewhere between one and two depending on whether the surface tension forces are dominant or the pressure differential forces are dominant. So, in the one case where the h dot term is very small compare one, you will get a dependence of linear dependence of the capillary force on r p. In the case where h star is much larger than one, you will get a r p square dependence, and for any value of h star, there is intermediate. You will get a dependence at somewhere between one and two

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If you look at the viscous forces on the other hand, there is clearly an r p squared dependence of the viscous force between two particles. Now, what the means is as particles gets smaller, the viscous forces will drop of much faster compare to the capillary forces; that is the normal viscous force. However, the tangential viscous force, which essentially relates to how particles are moving relative to each other in the direction of flow.

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Now, again when we say normal and tangential, what we mean is if your flow direction is like this. The normal force is the one that applies normal to the flow direction and the tangential force is what applies tangential to the flow direction. So, the viscous force is the normal viscous force goes as the square of particle size, whereas the tangential viscous force essentially goes as r p times logarithm of r p.

So, here again, the, if you look at the total viscous force and look at the particle size dependence, you will reach a value that is somewhere between one and two. Again depending on whether the tangential term is dominant or the normal term is dominant. Now, just like we had reported some imperial data on surface to particular adhesion which suggested a linear dependence on particle size as well as on the relative humidity that is presented in the system.

Similarly, that is quite a bit of work that has been done to look at agglomeration of particles in suspension from a purely empirical view point experimental. So, you take a bunch of particles, put them in suspension, monitor their number over time, total number of particles. As the total number decreases as a function of time, obviously there is more cohesion and agglomeration going on.

So, the rate of reduction in total number of particles as a function of time is a measure of the extent of cohesion that is present in the system. Another way to track it of course will be to look at size. If you track the mean size, that size will keep increasing as a function of time if there is cohesion.

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So, there are two ways in which you can detect the presence of cohesion or absence in a suspension and the both basically depend on plotting as a function of time. So, in one case, you can plot of t. So, let us say that this is time equal to 0 and the corresponding number of particles of any size. So, that is N at t equals to 0. What will happen is that if there is no agglomeration happening, then what a dependence would you expect? It will be constant right as a function of time.

So, if this is your N at t equal to 0, in the absence of cohesion, this number will remain constant over time. However, as cohesion starts to happen and as particle start to agglomerate, you will see that there is a net reduction in the number of particles as a function of time.

And similarly, you can also plot particle size d p as a function of time, and here again, if, let us say that this is your particle diameter at time equal to 0, let us say that this is your diameter at time equal to 0. If there is no cohesion, it will remain constant over time; whereas if there is cohesion that particle size will keep increasing in some fashion.

So, with cohesion, you will see and increase in the mean particle size of the system. So, this behavior, where there is a monotonic increase of particle size with time or a monotonic decrease in the number of particles with time is clearly indicative of cohesive phenomena that are happening in your suspension, and in fact, there is an empirical equation that governs the relationship between particle counts at various times.

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So, if you take 1 over N as at time t, this is equal to 1 over N at t equal to 0 plus 4 k t Cc t over three times mu - where N of t is the number of particles at time t. N at t equals to 0 is a number of particles at time equals to 0 plus k is a Boltzmann's constant; t is a temperature; lower case t is time; mu is viscosity and C c again is the stokes cunningham slip correction factor, and if you recall, I had mentioned that this is a roughly proportional to 1 over dp. So, effectively, you can write this or rewrite this as some constant k prime and have the particle diameter in the denominator. So, what is the implication of this equation? What is this tell us about agglomeration kinetics? What are the parameters that it depends on? Obviously, even if you take one over N of t minus one over N at equal to 0, this is now going to be equal to k prime k T t over three dp times mu.

So, if you examine this equation, which by the way as I said is empirical, what that means is people have thousands of experiments and actually recorded the data and they have done a regression analysis of the data to determine how particle count's change as a

function time and they have come up with this expression for various systems. What this tells us is that as temperature increases, there is increasing agglomeration; as time increases, there is increasing agglomeration; as particle size decreases, there is increasing agglomeration and that is why Nanoparticle systems in particular or very susceptible to cohesive behavior. There is a an excessive tendency for Nanoparticles to agglomerate in solution and form clusters, and finally, as viscosity decreases, there is an increasing tendency to agglomerate.

So, if you want to keep particles suspended and dispersed in solution, what this tells you is, actually it is gives you a lot of clues about how to keep particles separated, in, in suspension, use high viscosity liquids. So, is water at high viscosity liquid compare to some, but it is not the highest viscosity liquid. You can find; you can add additives to it to increase its viscosity. Particle size probably you cannot play too much; I mean you need Nanoparticles, you need Nanoparticles, right? However, temperature, now, that is interesting essentially by lowering the temperature, you can keep the particles more uniformly dispersed.

So, if you are trying to maintain a Nanoparticle suspension and ensure that the particle stay dispersed, what do you do? You freeze it; I mean that is a simplest thing to think of, because when you freeze it, the particles are now held where they are, right? And then just before you are ready to use the Nanoparticles, remelt the suspension. Alternatively, even if you do not freeze it lower the temperature, so that it is close to the freezing values and that will reduce the mobility of the particles, and thereby prevent agglomeration from taking place. Time obviously the longer you let the suspension set, the more tendency for cohesive behavior to happen.

So, if you want to use a Nanoparticle suspension, you use it as soon as you make it, or alternatively, you make a suspension just when you are ready to use it. So, it is like a just in time process. If you let any Nanoparticles suspensions sit for any length of time, you are going to see severe cohesive behavior. You will see particles disappear and you will see particles size is increase because of cohesion, agglomeration and so on.

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So, again, the interesting thing here is that when you look at extent of agglomeration, particle size it is in the numerator. Now, what that tells us is that the force of agglomeration between particles is actually scaling inversely with particle size, right?

So, this would tell you that F cohesion in general goes as 1 over dp, but if you look at the equations we have formulated, we are not really getting that trend anywhere, right? So far they are all either linearly dependent to particle size or linearly dependent on some power or exponent of particle size. So, how do you reconcile that with this behavior? That we know intuitively finer particles will find each other and adhere much faster compare to larger particles.

So, just like the, you know, the apparent conflict that we discussed earlier in terms of particle adhesion and particle removal, this is something else to kind of ponder. You know is there is there a theoretical reason for this? Can we explain this or is it completely anomalous behavior. So, let us we will come back this later.

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Ok. So, we have talked about Van der Waals forces, electro static forces, capillary forces and viscous forces. What do we mean by the spring forces? When you have two particles that are in suspension, they behave as if they are attached to a spring. There is a, in the normal direction, there is a compressive force associated with this pair of particles.

Over time as particles start moving away; let us say that particle one stays where it is but particle two has now moved over here. Then there is also a tangential spring force that applies F t, which is actually has a tendency to bring them back together. Both the F N force and the F t force are essentially attractive forces. The particles are constantly trying to, as you try to move them apart, they want to stick together, and so, they both lead to cohesive behavior. The higher the normal force and higher the tangential force, the greater will be the extent of cohesion.

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So, this force, the compressive force F n can be written as a k n times alpha to the power 3 by 2 - where alpha is dp 1 plus dp 2 minus s 1 2; where d p 1 and d p 2 are the diameters of the particles and s 1 2 is the distance of separation between the centers of the particles, and this parameter k n is given by 4 by 3 times d p star times square root of e star - where e is the young's modulus. d p star is calculated again by taking 1 over dp star equals 1 over dp 1 plus one over dp 2 and e star is calculated as 1 over e star equals 1 minus nu 1 square over e 1 plus 1 minus nu two squared over e 2 - where the nu value are poison's ratios.

So, if you look at the expression for the normal force, the key dependences are on the sizes of the particles. The distance of separation between the particles and on the associated parameters that govern the elastic behavior of the system, particularly, the poisons ratio and the young's modulus for the two particles.

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If you look at the tangential component, F t is related to F t 0 which is the time 0 value of the tangential force. That is basically where the two particles are virtually below each other; so, there is no tangential displacement, and then, as soon as the displacement starts to happen, this kicks in. So, it is actually a minus k t times delta s - where delta s is the tangential distance of separation between the two particles.

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Delta s of course is related to time delta t by the equation delta t times V t. So, if we take a time deferential delta t and multiplied by the tangential velocity, again the relative tangential velocity that gives you the distance of separation at time equals t. So, this is a relative value. So, basically the picture that you have here is when you have two particles that are adjacent to each other, they also a tendency to have a compressive interaction between them as well as a shearing interactions. So, this tangential interaction can also be written as a shearing force. But in this particular case, when you have two particles that are suspended in solution, the shearing force is one that essentially tries to bring them back together just as the normal force is a compressive force that tries to force the particles together.

So, again going back to the list we had, what you can see is virtually all of the cohesive forces are attractive in nature. Van der Waals forces are attractive; surface tension forces, pressure forces, viscous forces as well as the spring forces are all attractive in nature. The only force that is potentially repulsive is electro static forces. So, this again gives you a lot of clues about how to keep particles separated when they are suspended.

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Since all the forces of interaction, virtually all of them are attractive in nature. The first thing you have to understand is there is going to be a overwhelming tendency for the particles to attract each other. So, if you want them to repel each other, one way is to charge them. If you import a light charge on all the particles, then that F electrostatic can be made to be repulsive in nature.

So, let us sum up all the forces that act in cohesion. So, you have Van der Waal's plus electro static plus capillary plus viscous plus compressive plus shearing. So, of these, Van der Waals capillary, viscous, compressive, shearing are all attractive in nature. However, this can be attractive or repulsive. So, by inducing a large charge on the adjacent particles and ensuring that there of the same sign, you can setup a repulsive force which can balance or actually overwhelm all the contributions. So, if am trying to

minimize cohesive behavior, one of the techniques I would look at is using electro static effects to provide a repulsive force in the system.

What else can I do? Well, we know that, you know, these forces are attractive but you can make them weaker. For example, you have seen before that Van der Waals forces can be reduced by simply immersing in a liquid medium. So, if you are trying to again keep particles segregated, do not keep them as a dry powder. A dry powder has a much more severe tendency to agglomerate than wet slurry.

So, suspend the powder in a liquid so that you can minimize forces of Van der Waals attraction. Later on when you get ready to use the powder and you need it in dry form. You can always evaporate the liquid where they have suspended. Of course to do that, you are better of using a high volatility liquid which can be easily evaporated. Water is not an easy liquid to evaporate, but there are many organic solvents that you can suspend particles, in which, have a high volatility, and therefore, you can easily recover the particles in the dry state whenever you need them.

In terms of the capillary forces, surface tension is an important component. How do you minimize surface tension? By using surfactant and that is why the use of surfactant is again widely resorted to minimize cohesion in suspensions.

Now, there are two ways in which actually surfactants help - one is by reducing the surface tension forces and the other is by reducing the hydrophobic, hydrophilic interaction forces. By the way that we have not talked about here, but refer back to a discussion of interaction between particles and surfaces. The force of hydrophobic slash, hydrophilic interactions is another major contributor, and by the way, this force can also be made attractive or repulsive when you have particles in suspension.

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So, one way to reduce cohesive behavior of particle suspended, in a, in a liquid is that you have two particles and they are suspended in liquid and they have a high cohesive force between them. The use of surfactant helps in two ways - one is by reducing surface tension and reducing and reducing surface tension forces.

The other is if you can get that surfactant to actually coat on top of these particles, so, for example, simple non-ionic surfactant can be made to form a coating layer on the surfaces, then what will happen is the hydrophobic portion as we saw earlier in our discussion on surface interactions, the hydrophobic portion will attach itself to the particle. The hydrophilic portion will stick out.

Similarly here, the hydrophobic portion will attach to the particle and the hydrophilic portion will stick out, and these two will actually repel each other, because they both have a tendency to want to attach themselves to water, and therefore, surfactant help in two ways one is by reducing surface tension and the other is by providing an active coating on top of the particles, which can be made to provide again a repulsive interaction based on the hydrophobicity slash hydrophobicity of the surfactant layer compare to the medium of suspension.

So, you have to choose the surfactant appropriately for the liquid in which the particles are suspended. For example, if the suspension medium is water, then a simple non-ionic surfactant will work in this fashion.

But if the particles are suspended in a hydrocarbon solvent, then the same technique will not work, and in fact, in this case, the coating material should preferably be some low surface energy material like, for example, a fluorocarbon, because in that case, a fluorocarbon because it has low surface energy will reduce the force of interaction between adjacent particles.

So, the way that you are, you can reduce particle to particle cohesive forces is by the use of surfactants or detergents or solvents is in two ways - one is to affect the hydrophobicity and hydrophilicity and setup a repulsion mechanism. The other way is keep the attractive force but reduce its magnitude by reducing the surface energy of the particle by providing a suitable coating material.

So, if you have the surface that is coated with the fluorocarbon, a fluorocarbon by definition has very low surface energy which implies very low force of attraction. As we have discussed earlier, the surface energy of a material relates to its hamakar constant which relates its Van der Waals forces of adhesion. So, a fluorocarbon works by producing a low surface energy surface. A surfactant on the other hand works by lowering surface tension and by providing hydrophobic slash philic repulsion.

Let us come back to this equation. What else can be affect to minimize cohesive forces? We have talked about what we can do to minimize the Van der Waals force. We have talked about what we can do with the electro static force. We have talked about the capillary force. By the way, in terms of reducing capillary interaction, another way in which we can reduce the tendency to agglomerate is to reduce the initial concentration of particles. The lower the concentration of particles say N 0 value, the lower will be the cohesive phenomena that take place afterwards. So, in a in a highly dilute slurry or suspension, that tendency for cohesion the tendency for agglomeration will be significantly lower for many reasons. One of the simplest being the inter particle distance is much greater in a dilute suspension compare to concentrated suspension.

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So, it is more difficult for the particles to find each other and agglomerate. So, that gives us another option that if you want to maximize cohesion, increases the concentration in the slurry. If you want to minimize cohesion, you minimize the concentration of particles in the slurry.

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We talk about hydrophobic hydrophilic forces. Viscous force basically if you can reduce the viscosity, then that can actually result in reduced cohesion, compressive force and shearing force. Here essentially what you can do is - when you try to keep the particles apart, you know, the obviously the distance of separation plays a key role. The farther apart the particles are, the less is the compressive stress between them. So, this again says lower the concentration. If you reduce the concentration of particles, the inter particle separation will naturally be greater compare to if you have a more concentrated suspension.

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You can also look at parameters like the poison's ratio and the young's modulus and see how they can be tweaked in order to minimize these spring like forces, but in terms of the tangential or shearing force, essentially what this equation tells you is - if there is relative motion between the particles, there is a tendency for them to jerk back together.

Now, this actually gives us a clue as to why Nanoparticles have greater agglomeration tendencies, because Brownian diffusion which is the primary mechanism of transport for Nanoparticles has an inverse dependence on particle size. So, as particle size gets smaller, Brownian diffusion values get larger. So, if you have two adjacent particles, the distance by which they are separated as a function of time will be greater in the case where particles are smaller because they tend to move over larger distances.

And it is, since it is a random work, they are not going to move in the same direction. So, as particle size is gets smaller, this distance of separation delta s as a function of time will get larger.

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Basically this relative velocity V t will scale as 1 over dp. As the particle size becomes smaller and smaller, the relative velocity between adjacent particles will become larger, and therefore, the F t value will quickly increase, and so, the reason, the primary reason that particles in suspension have a greater tendency to attract each other. As the particle size gets smaller is this tangential or shearing interaction.

Now, there are other reasons as well and we will discuss them when we talk about the transport characteristics of particles, but the key thing to understand right now is that finer particles always have a greater tendency to its cohesion. Now, that has a lot of implications for again the flow ability of suspensions on slurries.

It is always more difficult to handle flows that involve fine particles compare to coarse course particles. So, we will stop this lecture at this stage. In the next class, we will talk about flow ability characteristics of suspensions. What are the properties of particles? Characteristics of particles that affect flow ability of suspensions and how they can be optimized for various applications. Any questions on what we talked about today? Ok. I will see you in the next class.