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The influence of particles on suspension rhec

Adding particles to liquids modifies the liquids' optical and physical properties, e.g. color, density, and viscoelasticit as water or acetone (https://wiki.anton-paar.com/us-en/acetone/), are typically ideally viscous liquids, while particle display a more complex rheological behavior. If the particles are soluble in the solvent, the final product is termed a Particles are dissolved as molecules or ions (<1 nm). In case the particles are insoluble, the product is a two-phase mixture which is termed a **suspension** if particles are larger than 1 µm or termed a **colloid** if particles are between size.

Many liquid products, paints, inks, beverages, medicine, slurries, or shower gels contain various types of particles desired final product or adjust processing properties. A low particle volume fraction (termed "solid fraction" hereafte shear-thinning behavior, whereas high particle concentrations might result in shear thickening. However, the mater not only depends on the concentration of particles but also on particle shape and particle size. In most suspension size, which can be expressed as the particle size distribution (https://wiki.anton-paar.com/us-en/particle-size-distrik Furthermore, the particle surface's electrical charge (zeta potential) influences the suspension's rheological behavi aims to separate the effects of particle concentration, particle shape, particle size distribution (https://wiki.anton-pa en/particle-size-distribution/), and particle surface charge, and to name common methods to measure these prope

Particles – the solid fraction in suspensions

Particles in a liquid act as obstacles, hindering the liquid's flow and therefore increasing the flow resistance, i.e. the simple model to describe the increased viscosity is to assume a flow field around a single sphere, representing ver concentrations, and an increased resulting viscosity simply calculated from the viscosity of the solvent and the volu dispersed solid (Figure 1, Equation 1, Einstein 1906, 1911).

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The formula was modified and adapted for multiple spheres of the same size ("monodisperse") for maximum solid **Products (https://www.anton-paar.com/us-en/products/)** 0.2 (Equation 2) by Batchelor (1977), with n representing the bulk viscosity of the suspension, η_0 the viscosity of the

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(https://wiki.antonpaar.com/fileadmin/wiki/images/influenceof-particles-on-suspensionrheology/fig1.png)

Figure 1: Flow around a single sphere in a suspension

Equation 1: $\eta = \eta_0(1+rac{5}{2}arphi)$

Equation 2: $\eta = \eta_0 (1 + 2.5 arphi + 6.2 arphi^2)$

Suspensions with an even higher solid fraction can be described with the model of Krieger & Dougherty (1959, Eq. low and high shear rates. In this model, two additional input parameters are required. Φ_{max} is the maximum solid fi suspension, still allowing the suspension to flow. The intrinsic viscosity [η] is a measure for the particle shape (2.5

Equation 3:
$$\eta = \eta_0 (1 - rac{arphi}{arphi_{max}})^{-[\eta] st arphi_{max}}$$



paar.com/fileadmin/wiki/images/influenceof-particles-on-suspensionrheology/fig2.png) Figure 2: Sphere stacking schemes with different maximum occupied volume solid fractions

Assuming spherical particles, the theoretical maximum packing density can be 0.52 to 0.74, depending on the sph scheme (Figure 2). It is well-established that for random close packing of spheres the value is about 0.64 (e.g. Scc

For low solid fractions, the viscosity of the suspension is increased. However, no significant particle-particle interac the flow behavior of the suspension is still controlled by the Newtonian flow behavior of the solvent phase. With inc fraction, it is more likely that particles collide. They now act as obstacles and additional shear force is required whe during particle collision (Figure 3). When the solid fraction approaches φ_{max} , ongoing particle-particle interactions increase the force required to shear the sample, now dominating over the shear stress required to shear the solver



(https://wiki.antonpaar.com/fileadmin/wiki/images/influenceof-particles-on-suspensionrheology/fig3.png)

Figure 3: Viscosity of sphere-bearing suspensions versus solid fraction for a constant shear rate and constant sphere size

The suspension's rheological response is now also shear-rate-dependent, the onset of non-Newtonian flow behavi thinning character of the suspension, is highly variable for different suspensions but may start at $\varphi = 0.1$ to 0.5 (e.g 2005). In addition, at higher shear rates particles form clusters and jamming occurs, leading to shear thickening (F transition from shear-thinning to shear-thickening is sample-specific and controlled by factors like particle size (dist particle shape.



(https://wiki.antonpaar.com/fileadmin/wiki/images/influenceof-particles-on-suspensionrheology/fig4.png)

Figure 4: Shear-thinning and shear-thickening behavior depending on the solid fraction of spherebearing suspensions

Particle size and particle size distribution

For a given solid fraction, smaller particle size leads to a higher number of particles in a given volume (Figure 5). T particle surface area can be several orders of magnitude higher for smaller particles. Very small particles display E acting against an applied shear force (Perrin 1910). Therefore, surface charge, adsorption, and hydration have a p effect on the effective hydrodynamic particle size for small particles in particular.



paar.com/fileadmin/wiki/images/influenceof-particles-on-suspensionrheology/fig5.png)

Figure 5: Different sphere size leads to different total sphere surface for a constant solid fraction

We have already shown that at a certain value of solid fraction, when particle-particle interaction becomes significa behavior is observed. The viscosity difference between small and large particles is diminished at higher shear rate favorably rearranged with respect to the flow direction (Figure 6). However, for the majority of all existing suspension of uniform and is best described as particle size distribution (https://wiki.anton-paar.com/us-en/particle-size-distrik Figure 7). The maximum packing density, and thus the maximum solid fraction, benefits from particles with variable ("polydispersity") as the space can be filled more efficiently.



(https://wiki.anton-

paar.com/fileadmin/wiki/images/influenceof-particles-on-suspensionrheology/fig6.png)

Figure 6: Shear-rate dependence of small and large spheres for a constant solid fraction. Both sphere sizes show shear thinning, although it is more pronounced for small spheres



(https://wiki.antonpaar.com/fileadmin/wiki/images/influenceof-particles-on-suspensionrheology/fig7.png)

Figure 7: Effect of particle size distribution (https://wiki.anton-paar.com/us-en/particle-sizedistribution/) on the maximum packing density for spheres. Polydisperse suspensions have a higher maximum packing density ϕ_{max} compared to monodisperse suspensions

In terms of the Krieger & Dougherty equation (Equation 3), φ_{max} increases (it may be above 0.74!) and thus reduce viscosity for a given particle concentration. The broader the particle size distribution (https://wiki.anton-paar.com/us distribution/) (PSD), the larger the positive effect on the packing density (Figure 7). We already learned that smalle a higher viscosity for a given shear rate and solid fraction. If we now compare the viscosity of two monodisperse al PSD, a viscosity minimum is evident for the polydisperse mixture (Figure 8). However, such polydisperse suspensi a high yield stress (Ancey & Jorrot 2001) and for colloidal suspensions with bimodal particle size distributions the s may act as a lubricant (Ancey 2001).



paar.com/fileadmin/wiki/images/influenceof-particles-on-suspensionrheology/fig8.png)

Figure 8: For a constant solid fraction and a constant shear rate, small spheres show higher viscosities compared to large spheres (compare Figure 6). Polydisperse suspensions have a higher φ_{max} . For a constant solid fraction (here: 0.63), φ / φ_{max} decreases and results in a viscosity minimum for such polydisperse suspensions.

Particle shape and surface properties

Until now we considered idealized spherical particles. Obviously, particles often display uneven convexity or non-s (elongation). The intrinsic viscosity [η] is a factor to describe the particle shape. For example spheres are represer 2.5 and ellipsoids with a 5:1, 10:1, 50:1 aspect ratio by values of 2.94, 3.43, 7.00 (Figure 9a).

For aspect ratios (R) from 1:1 to 20:1, the intrinsic viscosity can be expressed with the following function (Brenner 2006, Equation 4):

Equation 4: $[\eta] = 2.5 + 0.123(R-1)^{0.925}$

Irregularities of the particle surface lead to a higher viscosity for two reasons: The deflection of the flow lines of the around particles is stronger than for same-size spheres (Figure 9b), and the increased specific particle surface inc probability of particle-particle interactions (Figure 9c). The effect is stronger for high particle concentrations and high



(https://wiki.antonpaar.com/fileadmin/wiki/images/influenceof-particles-on-suspensionrheology/fig9.png)

Figure 9: (a) Aspect ratios of regular ellipsoids, (b) flow field around spheres versus flow field around irregular particles, (c) particle-particle interactions of spheres and irregular particles. Irregular particles show more contact points and thus a higher influence of inter-particle friction is expected.

If we now compare the shear-rate dependence of spheres and elongated particles, we recap that spheres show sł particle-particle interactions become less pronounced at high shear rates. The increased surface of elongated part higher low-shear viscosity compared to spheres. However, non-spherical particles have the possibility to adapt the flow direction. At rest, they are randomly oriented but align to the flow direction when a shear rate is applied. This I viscosity compared to spheres at higher shear rates because the intrinsic viscosity is then lower – resulting from th packing density φ_{max} (Figure 10).



(https://wiki.antonpaar.com/fileadmin/wiki/images/influenceof-particles-on-suspensionrheology/fig10.png)

Figure 10: Shear-rate-dependent viscosity of spheres compared to ellipsoids. Both show shear thinning. However, suspensions with ellipsoids have higher viscosities at low shear rates and suspensions with spheres have higher viscosities at higher shear rates.

The effect of electrical particle charge

Depending on the application of the suspension, a wide variety of substances are used for both solvents and partic having an electrical charge. The electrostatic momentum, which can represent attraction or repulsion within a susp quantified as zeta potential (https://wiki.anton-paar.com/us-en/zeta-potential/). If particles have a positive or negati molecules or ions with negative or positive charge occupy the surface (Figure 11) and together form a particle with surface potential.



(https://wiki.antonpaar.com/fileadmin/wiki/images/influenceof-particles-on-suspensionrheology/fig11.png)

Figure 11: Different types of particle-solvent interactions for a particle with negative surface charge: (a) no adsorption (high positive zeta potential (https://wiki.anton-paar.com/us-en/zetapotential/)), (b) adsorption and Stern layer formation (low positive zeta potential (https://wiki.anton-paar.com/us-en/zeta-potential/)) and (c) Stern layer and additional chemisorption (low negative zeta potential (https://wiki.antonpaar.com/us-en/zeta-potential/))

This electrical potential affects the interaction with the solvent as well as the inter-particle behavior. If the surface c repulsion between particles dominates over attraction by van-der-Waals forces (Figure 12a) and an energetic barri proximity of the particles. In contrast, for particles with near-zero surface charge, repulsion is weaker and an energe certain particle-particle distance facilitates an ordered particle arrangement (Figure 12b).



(https://wiki.antonpaar.com/fileadmin/wiki/images/influenceof-particles-on-suspensionrheology/fig12.png)

Figure 12: Particle-particle interaction as a function of inter-particle distance. (a) For high zeta potential (https://wiki.anton-paar.com/us-en/zeta-potential/), repulsion increases with decreasing inter-particle distance towards a maximum, represented by an energetic barrier. (b) For low zeta potential (https://wiki.anton-paar.com/us-en/zeta-potential/), agglomeration is favored by an energetic minimum at relatively small inter-particle distances.

In consequence, for a sample with a low zeta potential (https://wiki.anton-paar.com/us-en/zeta-potential/), accumul flocculation of particles is favored, whereas at a high zeta potential (https://wiki.anton-paar.com/us-en/zeta-potentia repel each other and effectively avoid agglomeration. However, to understand the rheological behavior of the susp additionally need to take into account particle size and solid fraction.

Dispersions with small particles (<1 µm) are particularly sensitive to changes in zeta potential (https://wiki.anton-papotential/) as induced particle repulsion and increased hydrodynamic volume (Maranzano & Wagner 2001, Figure the viscosity, in particular at low shear rates. This is only possible as gravitational forces act subordinate to such suparticles, while Brownian motion will dominate. Assuming relatively low solid fractions, this kind of dispersion will m shear viscosity plateau.



(https://wiki.antonpaar.com/fileadmin/wiki/images/influenceof-particles-on-suspensionrheology/fig13.png)

Figure 13: Effect of zeta potential (https://wiki.anton-paar.com/us-en/zeta-potential/) on a sphere-bearing suspension with constant solid fraction. The zero-shear viscosity plateau increases with increasing zeta potential (https://wiki.anton-paar.com/us-en/zeta-potential/).

In contrast, for larger particles the effect of sedimentation is generally stronger and together with the zeta potential paar.com/us-en/zeta-potential/) it will control the agglomeration behavior. At a low zeta potential (https://wiki.anton-en/zeta-potential/), particles stick together and by steadily increasing the size of the agglomerate, gravitation will fill sedimentation. Such agglomeration may induce a yield point if the solid fraction is sufficiently high and the particles three-dimensional network. Note the contrasting behavior at high shear rates, where shear thickening may occur, a repulsion causes higher viscosities for samples with a high zeta potential (https://wiki.anton-paar.com/us-en/zeta-p 14).



(https://wiki.anton-

paar.com/fileadmin/wiki/images/influenceof-particles-on-suspensionrheology/fig14.png)

Figure 14: For large particles (or higher solid fractions), low zeta potential (https://wiki.antonpaar.com/us-en/zeta-potential/) may lead to agglomeration and the presence of a yield point (example from Maranzano & Wagner 2001).

Methods to characterize suspensions

Bulk physical properties of the suspension and the solvent, like viscosity and yield point, can be measured with vis rheometers in rotational or oscillatory tests. Characterization of the solid fraction requires knowledge of particle siz distribution (https://wiki.anton-paar.com/us-en/particle-size-distribution/) (PSD), and particle shape. These propertie additional methods like dynamic light scattering or laser diffraction (for particle size and PSD) and high-resolution i particle shape) to be measured. Furthermore, electrophoretic light scattering is used to determine the zeta potentia (https://wiki.anton-paar.com/us-en/zeta-potential/).

Laser diffraction is able to analyze particles from the sub-µm to the mm size range. The measuring principle is bas laser directed to the sample and multiple detectors to record scattering at different angles. The light scattering beh respect to the particle size: smaller particles scatter more strongly than larger particles. However, the measuremer assumption of spherical particles and knowledge of the optical properties of the solvent as well as the particles is r

Dynamic light scattering allows particle size measurement down to the sub-nm range. The measuring principle use fluctuations over very short timescales due to Brownian motion. Hence, the particle speed can be correlated with p

For electrophoretic light scattering, two electrodes are connected to the sample and an electrical field is applied. C will now move towards either the positive or negative electrode, the velocity is measured with the laser Doppler tec frequency shift or as phase shift. The obtained values can be converted to zeta potential (https://wiki.anton-paar.cc potential/) distribution or an average zeta potential (https://wiki.anton-paar.com/us-en/zeta-potential/), respectively.

Automated imaging allows determination of particle shape and particle size if particles are at least 1 µm in diamete thousand images are taken to ensure statistically sound values. This method is particularly useful if particles show from sphericity. Except for dynamic light scattering (only applicable to wet samples), these methods can be used fc samples.

Conclusion

Many liquid products contain a certain fraction of particles and understanding of the viscoelastic behavior of these vital importance for a wide range of industrial, natural, and biological products and processes. Particle fraction, sha (distribution), and surface charge influence a suspension's rheological behavior, which is typically shear-thinning b Newtonian for low particle fractions, or is shear-thickening for high particle fractions at high shear rates. A combine determining the rheological properties and the particle properties is necessary for a comprehensive characterizatic

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