

Investigation of filtercake stability using numerical simulation technique

ANDERS SAND, MARTTI TOIVAKKA, AND TUOMO HJELT

ABSTRACT: The stability of a paper coating filtercake and the influence of Brownian motion relative to absorption flow can be described using a particle dynamics model. For such a study, we assumed that the stability of a filtercake depends on the interrelation between particle diffusion and the hydrodynamic drag force caused by the absorption flow of liquid. We simulated particle diffusion using a Brownian motion model. Our investigation included monodisperse and polydisperse particle systems of electrostatically stabilized rigid spheres. We also varied system temperatures in some of the simulations. Results characterize the stability, microstructure, density profiles, and thicknesses of the filtercake. We concluded that particle diffusion disrupted the filtercake if the size of the particles was small or the temperature high. For dewatering rates typical of paper coating processes, the simulations show that only particles less than 0.2 μm are able to diffuse sufficiently to result in a solids density gradient over the thickness of the filtercake. In the polydisperse system, small particles can migrate within the filtercake at low dewatering rates. The solids concentration gradient observed in some cases appeared to be the result of filtercake surface roughness, partly caused by a size segregation effect rather than of the proposed thickening mechanism.

Application: This paper describes a novel approach to investigating the structural formation and stability of paper coating filtercakes. Results help to further our insight into fundamental physical phenomena related to pigmented coatings.

On application, coating colors penetrate the fiber structure of base paper. In addition, a dense layer of pigment particles, called *filtercake*, forms at the coating suspension-paper interface during initial application [1-3]. The structure of this filtercake will partly influence the final structure of the fully immobilized coating layer. It therefore also determines the physical and functional properties of the coated paper. For example, the absorptive properties of the coating layer affect printability, while the distribution of binder will influence its strength properties. Filtercake formation also contributes to the runnability of a coater.

The accumulation of pigment particles at the base substrate has been investigated both empirically and theoretically over the years [1, 2, 4-6]. This complex mechanism, being a result of pressure-induced penetration or capillary absorption of liquid phase into the base paper, has been the subject of long-standing disputes within the research community. Several research groups have used modeling and simulation to investigate the potential mechanisms that govern the dynamics of the layer formation [7-11]. While most studies have focused on the accumulation of particles, this work is aimed at studying mechanisms that have an influence on the already formed layer, thereby characterizing its dynamic behavior and stability.

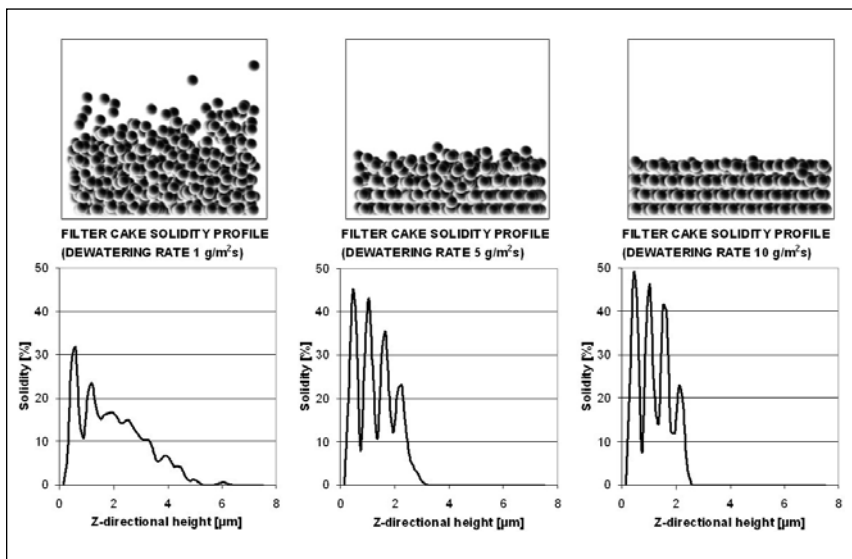
Two dominating theories have been proposed to explain the mechanism of filtercake formation:

- Immobilization through filtercake formation assumes a sharp solids concentration difference between the filtercake and the bulk coating suspension.
- Immobilization through thickening supports the existence of a rather low concentration gradient throughout the filtercake structure.

These mechanisms have been debated in many literature sources [4]. Engström [5], for example, observed a filtercake concentration gradient in his pilot trials and Lohmander et al. [4], as a result of their laboratory scale experiments, reported the solids concentration decreases to be nearly linear as function of distance from the filter medium. Work by Salminen et al. [6], however, did not support the existence of this concentration gradient. The theories are of importance in many theoretical calculations on filtercake properties. In contrast to thickening, the filtercake formation mechanism assumes a nearly incompressible filtercake, which is the basis of traditional permeability calculations based on Darcy's law.

Varying conclusions reported in the literature can be explained by the complexity of measurement techniques and the differing experimental setups that have been used. Experimental methods range from destructive scraping techniques to magnetic resonance imaging [4-6]. All exhibit shortcomings that potentially influence the results. The application of numerical simulation in investigating mechanisms that govern pigment particle behavior on the colloidal level has proven to be promising in many ways [12-14]. Computer simulation of coating particles enables direct observation and identification of potential behavioral mechanisms and estimates of properties that are difficult or impossible to determine empirically.

The model used in this work uses a modified Stokesian dynamics simulation technique. It describes the microscopic movement of particles and the constitution of particle systems by calculating interparticle hydrodynamic and DLVO-type (Derjaguin-Landau-Verwey-Overbeek) colloidal interactions. In this work, we assume that the structure and stability of the



1. Filtercake structures at three different dewatering rates (situation at 280 s). Simulation pictures (above) visually illustrate the solidty profiles (below).

filtercake will be determined by the ability of particles to diffuse against an absorption flow of liquid.

METHODOLOGY

The filtercake stability investigations were performed using the particle dynamics code PD3D developed by Toivakka and Nopola at Åbo Akademi University [15, 16]. The simulation technique is based on Stokesian dynamics [17, 18], which is capable of predicting the motion of particles suspended or dispersed in a liquid medium, making it suitable for simulating the behavior of coating colors. The technique is, however, computationally intensive and without modifications limited to small systems of a maximum of a few hundred particles. Several alterations have been made by different research groups to the original Stokesian dynamics method to increase computational effectiveness. These include the lubrication approximation, introduced by Bousfield, which estimates interparticle hydrodynamic interactions by near-field lubrication forces and a Stokes drag contribution from the bulk flow field [19]. The lubrication approximation was later modified by Toivakka [15]. The applicability of the approximation is confined to concentrated suspensions, but for coating colors with typically high solids volume concentrations this is not a limitation. Further modifications include the sparse matrix and neighbor list techniques, which have

earlier been reported by Toivakka (1997) and Nopola (2004) [15, 16].

For this work, the existing simulation software was modified by including a novel Brownian motion model based on the Einstein diffusion equation [20]. That provided a means to study diffusion and particle redistribution phenomena caused by thermal vibration of particles, random collisions with molecules of the suspending medium, other particles and system boundaries. Although being of relevance for all suspended particles of a coating color, including pigment particles and polymeric material, the influence of diffusion is thought to be of importance mainly for small coating particles and additives, particularly binders (100-300 nm) and thickeners (5-10 nm).

The main functional principle of the Brownian motion model is to apply a force of random direction to each particle at each time step. The R250 random number generator [21] was used in generating a randomly directed 3-D unit vector (e.g., the trigonometric algorithm described by Allen and Tildesley [22]) to represent the direction of the force. The model was tested and showed good compliance with analytical sources [20, 23]. Details of the model can be found in Appendix I. The base substrate is approximated as a completely planar and smooth surface with uniform dewatering rates, i.e., without any local variations in absorption or roughness. A

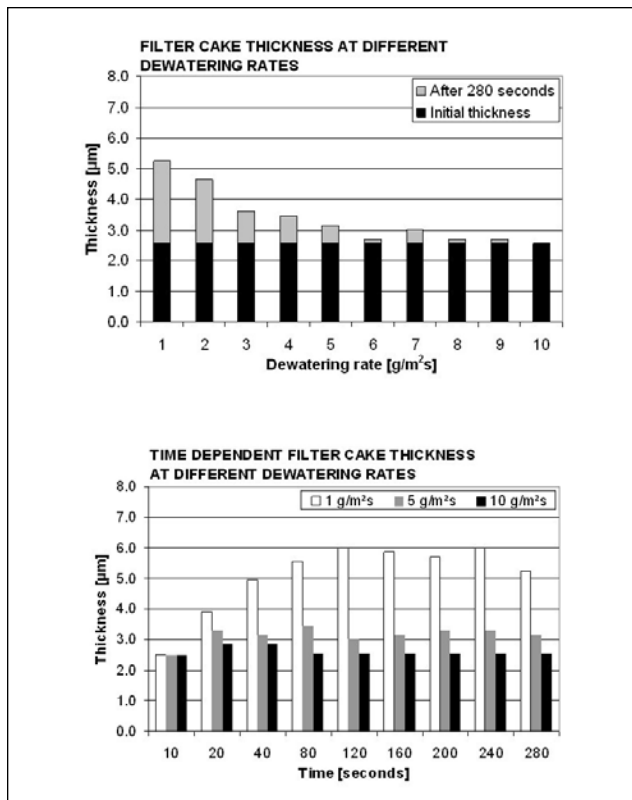
mechanism that has also been proposed for settling or consolidating systems under shear is a phenomenon termed shear-induced particle resuspension [24]. This work does not, however, assume any lateral-direction flow of liquid across the filter cake. In some applications, cross-direction flow of liquid has been shown to be a probable cause for particle migration.

SIMULATION CONFIGURATION

Filtercake structural properties and stability were studied as a function of base substrate absorption rate, particle size, and size distribution. We also examined the potential influence of thermal conditions by accounting for temperature in the Brownian motion model and by adjusting liquid phase viscosity accordingly. Unless specifically stated, the temperatures of the systems were set to 20°C. DLVO-type colloidal interactions were not of interest in this study, but some electrostatic repulsion (double layer thickness, $1/\kappa = 5$ nm) was applied to avoid excessive particle clustering effects. Note that the colloidal interaction model parameters used in this work do not promote flocculation, which would reduce the particle mobility. Flocculation would influence filtercake formation, its density profile, and possibly particle diffusion away from the filtercake.

Monodisperse and polydisperse systems of spherical particles, ranging between 0.2 μm and 2 μm , were the subjects of investigation. Particle sizes were chosen so that the smallest particle size would correspond to the approximate size of binders, while the largest particles would be roughly the same size as relatively large ground calcium carbonate (GCC) pigment particles. For finer commercial GCC grades, this size range would account for about 80% of the solids volume. The particle size distribution of the polydisperse system was consequently adapted to correspond to commercial GCC (Hydrocarb™ 90, Omya, Switzerland).

The base substrate was approximated as a completely planar boundary with absorption rates dimensioned to be in the scale of 1 $\text{g/m}^2\text{s}$ to 10 $\text{g/m}^2\text{s}$. This



2. Relationship between filtercake thickness and dewatering rate (top) in a system of 0.5 μm monodisperse particles. The increase in filtercake thickness as function of time (bottom) indicates diffusion.

study does not back couple base substrate absorption rate to filtercake permeability properties. In experimental conditions, this could be compared to keeping dewatering rates constant by applying external pressure to the system. Since the liquid flow caused by base substrate absorption was estimated to dominate, gravitational forces were not included in the simulations.

Filtercakes were formed by allowing an initially high dewatering rate of 25 g/m²s to force all particles of the simulated system to form a dense filtercake. After the initial formation of the filtercake, the dewatering rate was reduced to a simulation-specific set value between 1 g/m²s and 10 g/m²s, based on empirical experiences [2], whereby the stability of the filtercake could be investigated.

Note that absolute filtercake thickness is not relevant to this study, since particle quantity (solids content) and system size were tailored only to promote comparability between simulations. The solids content and subsequent filtercake thickness were, however, selected to roughly correspond to that which can be expected in practical coating processes.

RESULTS & DISCUSSION

Influence of dewatering rate

We investigated several monodisperse systems, focusing on the influence of dewatering rate and particle size on filtercake stability. The influence of the dewatering rate was stud-

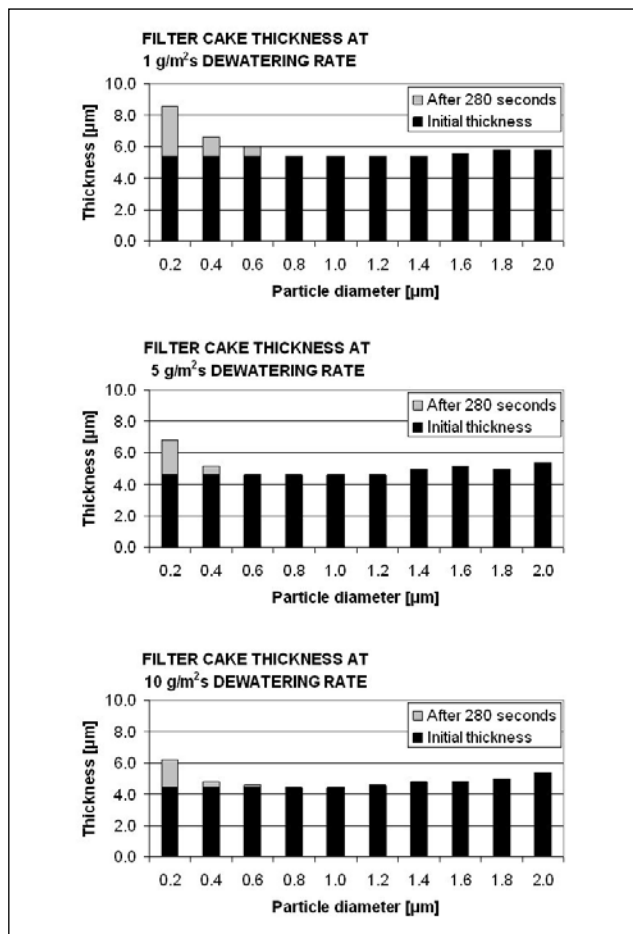
ied in a system containing 500 0.5 μm particles. **Figure 1** illustrates the simulation output at $t = 280$ s, in which each peak of the solidity graphs represents an ordered layer of particles. At a dewatering rate of 10 g/m²s, particles were arranged into a distinct structure of a few monolayers, while enabling small structural irregularities to be introduced if the dewatering rate was lower. At the lowest dewatering rate, single particles or groups of particles were able to completely diffuse into the bulk liquid phase as shown by the leftmost picture in Fig. 1.

The stability of the filtercake was characterized by comparing its initial thickness with the thickness at different simulation times. Filtercake thickness was defined as the z-directional height above the absorbing base substrate where there would still be a continuous phase of solid material, i.e., completely diffused individual particles were defined as not being part of the filtercake. **Figure 2** gives a rough approximation of diffusion by comparing initial and final filtercake thickness. In fact, a steady-state value of filtercake thickness could be located, where either the filtercake was stable or particles of the top layer would be able to completely diffuse away from the structure. This thickness was found to be intimately related to the dewatering rate. Keep in mind, however, that in the event of particle diffusion into the bulk of the liquid, filtercake thickness would slowly decrease as a result of its gradual deterioration.

Influence of particle size

Monodisperse systems of different particle sizes, ranging between 0.2 μm and 2 μm , were designed to investigate the influence of particle size on the stability of the filtercake. We selected three different dewatering rates—1, 5, and 10 g/m²s—to study particle diffusion against liquid flow. The particle system sizes were planned to achieve equivalent solids concentrations and roughly the same filtercake thicknesses, given a certain packing ratio. Some artifact effects in the filtercake thickness could be observed in the systems with the largest particle diameters, due to the few particle monolayers in the filtercake. The number of particles in all systems was set to 1000.

Simulation results, illustrated in **Fig. 3**, indicate that only small particles, i.e., particles with diameters of 0.6 μm or less, are sufficiently mobile to diffuse and cause filtercake deterioration in the specified flow fields. Although being less pronounced at high base substrate absorption rates, the diffusion capability of the 200 nm particles appeared relatively similar at all dewatering rates investigated. This was quite in contrast to the somewhat larger (400-600 nm) particles. Their diffusion layer (marked grey in Fig. 3), quantitatively defined as the filtercake thickness difference between the initial situation and the situation after 280 s, appeared to decrease faster as a function of the dewatering rate relative to what was the case in the 200 nm particle system. This observation led to the conclusion that the extremely high mobility of small particles makes their diffusion capability less dependent on liquid flow rate. The



3. Relationship between filtercake thickness and particle size, studied at three different dewatering rates. The difference between initial thickness and thickness after 280 s indicates filtercake deterioration caused by diffusion.

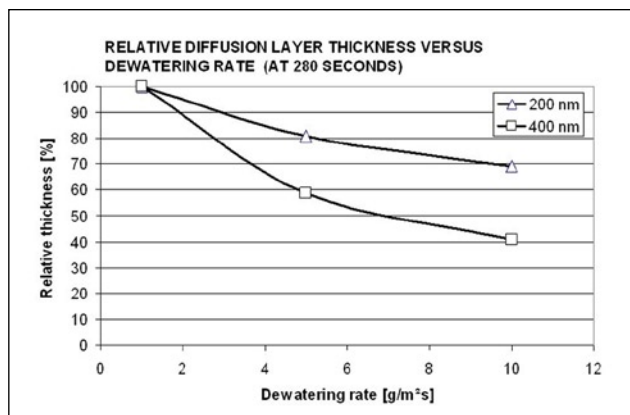
mobility of the 400–600 nm particles was consequently found to be more dependent on the dewatering rate.

This phenomenon is illustrated in **Fig. 4**, in which the 200 nm and 400 nm particle systems are compared. The system with the larger particle size, 600 nm, was omitted from **Fig. 4** because it exhibited enough quantitative uncertainty to preclude any valuable comparison with the other systems. Relative thickness was defined as the diffusion layer thickness as a fraction of that of the 1 g/m²s dewatering rate for the specific system. Thereby, the thickness curves were made comparable by being normalized to 100% at the slowest dewatering rate.

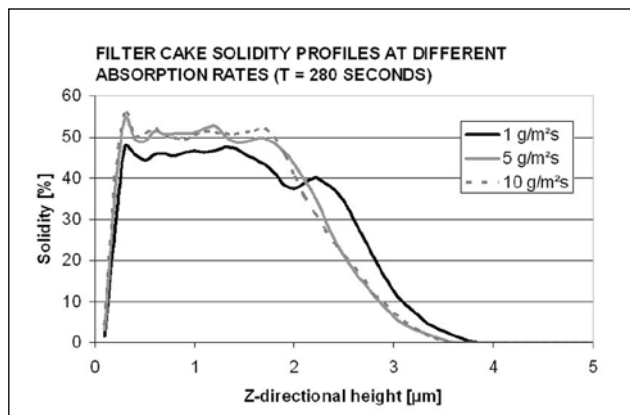
Polydisperse systems

The polydisperse system was designed to have a particle size distribution comparable to commercial GCC. We simulated 1600 particles, with sizes ranging from 0.2 µm to 2 µm in 0.05 µm to 0.10 µm increments. Consequently, particles of 26 different sizes were simulated in the same system.

In contrast to the monodisperse systems, the filtercake solidity profile (**Fig. 5**) appeared fairly static in the polydisperse system, regardless of the dewatering rate. This can be explained by the low diffusion capability of large pigment particles in combination with their distribution in the filter-



4. Reduction in diffusion layer thickness at increasing dewatering rates. The mobility of smaller particles appears to be less restrained by dewatering rate.



5. Solidity profiles for the polydisperse particle system after 280 s of simulation time. Filtercake structures were compared at three different base substrate absorption rates.

cake. Consequently, the presence of large particles appeared to impede free movement of otherwise extremely mobile small particles within the filtercake, preventing any significant diffusion from taking place. The phenomenon of size segregation, previously described by Toivakka et al. [14, 15] and observed in similar simulations [8–10], was concluded to be more pronounced at higher dewatering rates, **Fig. 6**.

In this context, however, size segregation could be interpreted as the inability of small (200–400 nm) particles to migrate through the filtercake pore structure against any significant liquid flow rate. The resulting thickness of the filtercake therefore remained relatively constant. With some minor internal redistribution of fine particles being the only detectable mechanism for structural change in the filtercake during the time interval investigated, the only external sign of diffusion was a slight change in the top layer concentration gradient.

Due to the particle size distribution, no systematic layering could be observed, as was the case in the monodisperse systems. The density remained relatively constant throughout the bulk of the filtercake. The densities were somewhat higher for filtercakes subjected to a higher dewatering rate.

COATING

Influence of temperature

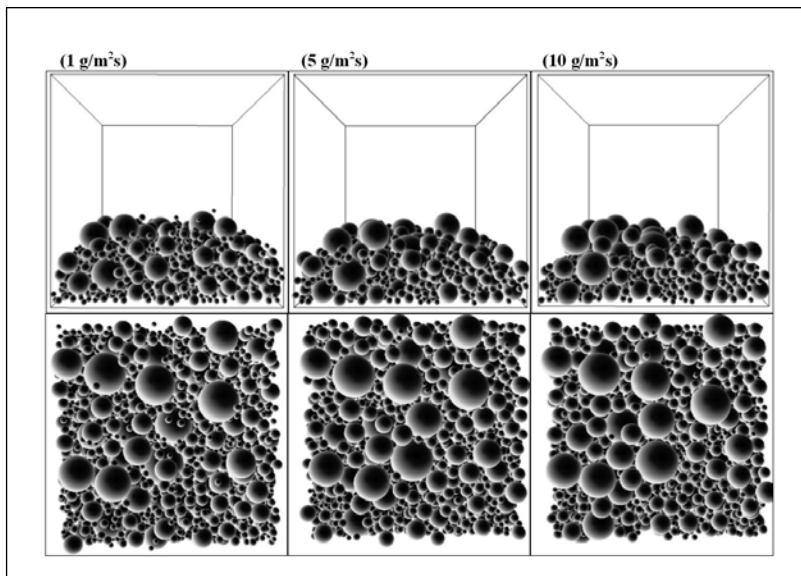
The influence of temperature on particle mobility was studied in a few simulations of monodisperse particle systems. The thermal influence was taken into account by adjusting the temperature parameter in the Brownian motion model between 20°C and 80°C. We also modified the liquid phase (water) viscosity in accordance with its temperature dependence, both in the Brownian motion and hydrodynamic interaction models. Simulations were thereafter run with temperature as the only changing variable.

We expected to see a significant effect on particle diffusion properties, due to the increased tendency for thermal motion along with reduced liquid phase viscosity, but thermal conditions appeared to play a less central role than initially anticipated. Some comparisons of filtercake solidity profiles at different system temperatures are shown in **Fig. 7**. The most extreme case, with the smallest particle size and slowest dewatering rate, shows clear differences in filtercake structure as a result of the higher system temperature. With slightly larger particles and a higher dewatering rate, however, this effect is barely observable. By adjusting thermal conditions in systems of small particles, it is possible to influence the concentration gradient. On the other hand, it would be quite interesting to further clarify the influence of temperature on the distribution of binders and thickeners in the coating layer due to their influence on coating layer mechanical and optical properties.

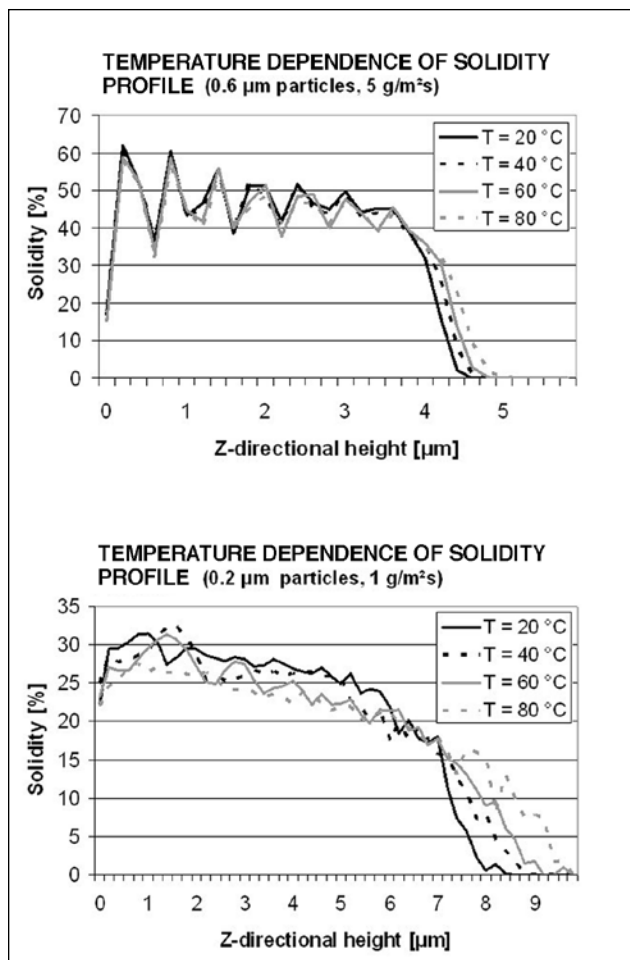
SUMMARY & CONCLUSIONS

At slow dewatering rates, which can be considered typical for laboratory scale experiments, diffusion of particles from the filtercake promotes the formation of a sometimes close to linear concentration gradient. This gradient, however, quickly becomes steeper at increased dewatering rates, which can be explained by the inability of particles to diffuse against the liquid flow. At dewatering rates of around 5 g/m²s, the boundary is already fairly sharp for most particle sizes. Consequently, at coating layer immobilization times typical for pilot and full scale coating processes [25], the results of the simulations indicate that the filtercake-coating suspension interface can be expected to be fairly sharp and that any significant migration of particles is unlikely.

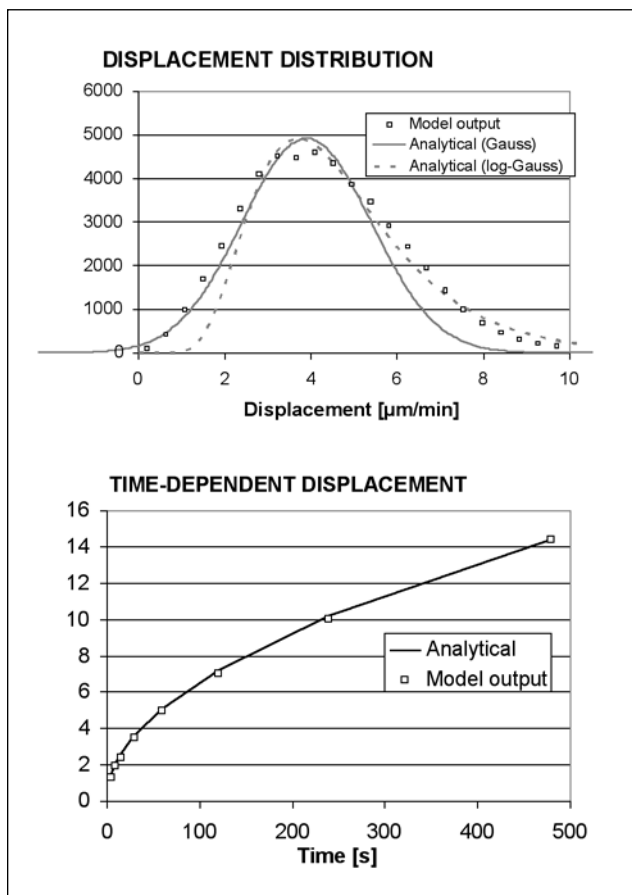
In the polydisperse system, apparent size segregation was observed as a function of increased dewatering rate. The migration of small particles within the filtercake was detected by small differences in the filtercake solidity profiles. Due to the size segregation phenomenon, it appeared relatively restrained by the existence of large immobile particles. The concentration gradient observed in the polydisperse systems resulted from filtercake surface roughness rather than any thickening or thinning type of formation or degradation mechanism.



6. Side (above) and top (below) view of the filtercake structure of a polydisperse particle system at absorption rates of 1, 5, and 10 g/m²s; situation at 280 s of simulation.



7. Influence of thermal conditions on filtercake solidity profiles (situation after 280 s of simulation).



8. Comparison between particle displacement distribution generated by model and the analytical Gaussian and log-Gaussian distributions (top) and time-dependent particle displacement compared to the analytical Einstein equation (bottom).

Although temperature affects the viscosity of the coating color and diffusion has been shown to be temperature dependent [20], this study concludes that the influence of thermal conditions is fairly negligible in this context. This study does not, however, take into account how the absorptive properties of the base substrate are related to temperature, although earlier publications have considered that relationship [1].

APPENDIX I

A Brownian motion model was included to describe diffusion and particle redistribution phenomena caused by thermal vibration of particles, random collisions with molecules of the suspending medium, other particles, and system boundaries. The influence of diffusion is thought to be of importance mainly for small coating particles and additives, e.g. latex (100-300 nm) and thickeners (5-10 nm), although being relevant also for larger particle sizes. Furthermore, a higher system temperature promotes diffusion through increased mobility of molecules and the reduced viscosity of the liquid phase.

The model is based on the well-known Einstein equation for calculating the mean Brownian displacement, $\bar{x}$, of a particle as function of time, t , as

$$\bar{x} = \sqrt{2Dt} ,$$

where particle diffusivity, D , is a function of temperature, T , viscosity, η , and particle radius, a , according to the equation

$$D = \frac{RT}{6\pi\eta a N_A}$$

where R is the universal gas constant and N_A Avogadro's constant [20].

The general functional principle of the Brownian motion model is to apply a force of random direction to each particle at each time step. For simplicity, the force will be constant for a specific particle size and system temperature. While increasing computational time and complexity, there would not be any particular benefit in calculating a force of both random magnitude and direction. Also in an effort to increase computational efficiency, the R250 random number generator [21] was used in the generation of random numbers.

There are several methods of creating a random 3-D unit vector to represent the direction of the force [22]. In this model, the direction is determined by the trigonometric method of generating uniformly distributed points on a unit sphere. This takes place according to the following algorithm:

- Choose z uniformly distributed in $[-1, 1]$.
- Choose t uniformly distributed in $[0, 2\pi]$.
- Let $r = \sqrt{1 - z^2}$.
- Let $x = r \cos(t)$.
- Let $y = r \sin(t)$.

When the unit vector $\bar{a}^0$ has been determined by the algorithm, the random force can be calculated as

$$\bar{F} = K_{Brown} \bar{a}^0 \sqrt{a} ,$$

where a is particle radius and K_{Brown} is an iteratively determined, system-specific constant. K_{Brown} is iteratively determined from calibration data, relating its value to the desired system temperature and liquid phase viscosity of each individual simulation. The model was tested and showed good compliance with analytical sources. The histogram of $n = 49,900$ random displacements of an $r = 1.7 \mu\text{m}$ particle is presented in **Fig. 8** (top), where the model output is compared with the analytical distributions. At the bottom in Fig. 8, the displacement of an $r = 1.0 \mu\text{m}$ particle is compared with the analytical results of the Einstein diffusion equation.

ACKNOWLEDGEMENTS

This work is part of the KCLCONS project "Coating Mechanisms" coordinated by Oy Keskuslaboratorio - Centrallabo-

ratorium Ab (KCL). We also acknowledge the National Technology Agency of Finland, TEKES, for financially supporting the project.

Received: June 16, 2006
Accepted: March 11, 2007

LITERATURE CITED

1. Eklund, D.E. and Salminen, P.J., *Tappi J.* 69(9): 116(1986).
2. Letzelter, P. and Eklund, D., *Tappi J.* 76(5): 63(1993).
3. Letzelter, P., "Studier över avvattningen av en bestrykningssmet i en bladbestrykare," Licentiate thesis, Laboratory of Paper Chemistry, Åbo Akademi University, Turku, Finland, 1993.
4. Lohmander, S., Martinez, M., Lason, L., et al., *J. Pulp Paper Sci.* 27(6): 183(2001).
5. Engström, G., *Wochenblatt für Papierfabrik*, 114(66): 195(1986).
6. Salminen, P., Roper, J., Pollock, M., et al., 1995 TAPPI Coating Conference Proceedings, TAPPI Press, Atlanta, Georgia, USA, pp. 277-286.
7. Eksi, G. and Bousfield, D.W., *TAPPI J.* 80(2): 125(1997).
8. Vidal, D., Zou, X., and Uesaka, T., "Modeling coating structure development: Monte-Carlo deposition of particles with irregular shapes," Proceedings of the 2003 TAPPI 8th Advanced Coating Fundamentals Symposium, TAPPI PRESS, Atlanta, CD-ROM.
9. Vidal, D., Zou, X., and Uesaka, T., *TAPPI J.* 86(4): 3(2003).
10. Vidal, D., Zou, X., and Uesaka, T., *TAPPI J.* 86(5): 16(2003).
11. Hiorns, A.G. and Nesbitt, T.E., "Particle packing of blocky and platey pigments—a comparison of computer simulations and experimental results," Proceedings of the 2003 TAPPI 8th Advanced Coating Fundamentals Symposium, TAPPI PRESS, Atlanta, CD-ROM.
12. Toivakka, M., "Numerical investigation of droplet impact spreading in spray coating of paper," Proceedings of the 2003 TAPPI 8th Advanced Coating Fundamentals Symposium, TAPPI PRESS, Atlanta, CD-ROM.
13. Toivakka, M. and Bousfield, D.W., 2001 TAPPI Coating Fundamentals Symposium Proceedings, TAPPI PRESS, Atlanta, p. 293.
14. Toivakka, M. and Eklund, D., *Tappi J.* 79(1): 211(1996).
15. Toivakka, M., "Simulation of particle motion in pigment coating colors," PhD thesis, Laboratory of Paper Chemistry, Åbo Akademi University, Turku, Finland, 1997.
16. Nopola, T., "Simulation of Particle Motion in Concentrated Colloidal Suspensions," Licentiate thesis, Department of Mathematics, University of Turku, Turku, Finland, 2004.
17. Brady, J.F. and Bossis, G., *J. Fluid Mech.* 155: 105(1985).
18. Brady, J.F. and Bossis, G., *Annual Review of Fluid Mechanics* 20: 111(1988).
19. Bousfield, D.W., 1990 TAPPI Coating Conference Proceedings, TAPPI PRESS, Atlanta, pp. 325-334.
20. Shaw, D.J., Introduction to Colloid and Surface Chemistry, 4th edn., Butterworth Heinemann, Oxford, UK, 2003.
21. Vattulainen, I., Ala-Nissilä, T., and Kankaala, K., *Physical Review Letters*, 73(19): 2513(1994).
22. Allen, M.P. and Tildesley, D.J., Computer Simulation of Liquids, 1st edn., Oxford Science Publications, New York, USA, 1987.
23. Sharma, N. and Patankar, N.A., *J. Computational Physics* 201: 466(2004).
24. Shauli, A., Wachs, A., and Nir, A., *Int. J. Multiphase Flow* 26: 1(2000).
25. Timofeev, O., Pajari, H., Koskela, H., et al., "Drying of coated paper: simulation and pilot experiments," PAPTAC 93rd Annual Meeting, PAPTAC, Montreal, Canada, 2007.

INSIGHTS FROM THE AUTHORS

The formation and structure of the filtercake in consolidating pigment coatings has been a subject of keen interest for many years. The conclusions of experimental and theoretical studies have been ambiguous, suggesting two opposing immobilization mechanisms: filtercake formation vs. thickening. We believe that the divergent results of the empirical studies on the subject are partly due to the different experimental setups and approaches used. An interesting conclusion of our work was that both mechanisms could be observed, depending on experimental boundary conditions and particle system properties.

This research complements previous work by focusing on the stability of already formed filtercakes at different conditions, instead of studying their formation. Numerical simulation constitutes a valuable approach to investigating the dynamics of filtercake mechanisms without many of the restraints of experimental studies.

Despite advances in computer technology, insufficient computing resources continue to challenge the numerical modeling research. Making a simple enough model that still portrays the key elements of the phenomena under investigation remains an integral part of the modeling scientist's work.

This study provides input to the understanding of



Sand



Toivakka



Hjelt

coating process mechanisms. Continuous efforts are made to develop the models and simulation techniques, which will prove to be an increasingly sophisticated tool in exploring fundamental aspects of pigment coating and guide papermakers in the choice of optimal process parameters.

Sand and Toivakka are with Laboratory of Paper Coating and Converting, Åbo Akademi University, Turku, Finland; Hjelt is with KCL, Espoo, Finland; email Sand at anders.sand@abo.fi.