

The influence of kaolin shape factor on the stiffness of coated papers

J.C. Husband, L.F. Gate, N. Norouzi and D. Blair

TAPPI Journal June 2009 pages 12-17

⊕ KAOLIN

⊕ GCC

⊕ PCC

The influence of kaolin shape factor on the stiffness of coated papers

J.C.Husband, L.F.Gate, N.Norouzi[†] and D.Blair[†].

Pigments for Paper Group, Imerys Minerals Ltd., Par Moor Centre, Par, United Kingdom PL24 2SQ,
[†]Dept of Chemistry, University of Edinburgh, King's Building, West Mains Road, Edinburgh EH9 3JJ,

ABSTRACT

The mechanical properties of paper coating layers are very important in converting and printing operations. These include stiffness, resistance to fold cracking, dusting and pick resistance. In this study we have prepared unsupported coating layers and measured the in-plane tensile properties as a function of the kaolin particle aspect ratio. We have then compared the results to pilot coated papers containing the same clays.

Kaolin – based coating layers differ from those based on ground calcium carbonate because of the platy nature of the clay particles. This gives different mechanical properties in the x-y and z-direction. We have shown that a coating layer containing high aspect ratio fine clay has an elastic modulus 7-8 times higher than one containing only ground calcium carbonate. With appropriate binder systems, these layers can have a modulus which is of the same magnitude as that of the basepaper. We show that this improvement can be used with advantage to increase stiffness or decrease basis weight whilst maintaining stiffness, in order to save costs.

INTRODUCTION

Among the mechanical properties of coated papers, stiffness and strength are perhaps the most important. The surface strength of coated paper is critical to the printing process. Resistance to cracking is important during the converting of paper and board. As postal rates increase, reduction in the basis weight of publication papers is desirable, but maintaining the stiffness of coated papers is paramount. It is interesting that reduction of basis weight from 30 to 28 lb ream⁻¹ for a major US title was unsuccessful because the magazine had an “insubstantial feel” [1].

Kajanto [2] has written an introduction to the bending resistance of paper. For a sheet of uniform composition in the z-direction, bending stiffness is shown to be related to the elastic modulus times the cube of the thickness (1) :

$$S_b = Ed^3 / 12.....(1)$$

The presence of multiple layers such as in a coated paper, for example, affects the stiffness according to the moduli of each layer, their thickness, and position relative to the neutral plane of the paper. The simplest case is that of a symmetrical three layer structure such as a two-side coated (2CS) sheet shown diagrammatically in Figure 1. In this case the stiffness of the sheet is given by :

$$S_b = \frac{E_2 d_2^3}{12} + \frac{E_1 (d^3 + d_2^3)}{12}(2)$$

where E_1 and E_2 are the elastic modulus of the coating and basepaper layers respectively, and d_2 and d are the thickness of the basepaper and coated sheet, assuming equal coat weights on each side.

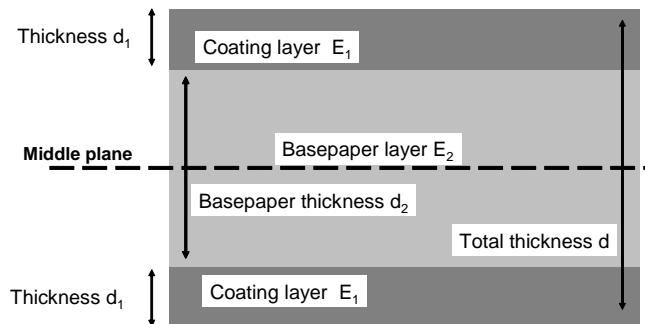


Figure 1. Representation of a two-side coated sheet.

If high stiffness is being sought, equation (2) predicts that it is advantageous if the coating layers have a higher modulus than the basepaper. This is not normally the case. Therefore measures which increase coating layer modulus should be beneficial for stiffness.

A number of investigators have studied the mechanical properties of coating layers. Parpillon *et al.* [3] measured the tensile strength of coating layers based on kaolin and calcium carbonate (GCC). They found that, at the same latex level, kaolin gave coating layers with about twice the tensile strength of GCC. Kaolin also gave greater stiffness. Subsequently, Lepoutre and Rigdahl [4] examined a wider range of pigments of different shape and related the in-plane elastic modulus of coating layers to the presence of voids. Further studies were reported by Inoue and Lepoutre [5]. They concluded that coating layers based on plate-shaped clay particles were more resistant to in-plane stresses than isometric GCC.

Engineering the particle shape, or aspect ratio, has made kaolin a very versatile coating pigment which confers additional functionality to coating formulations [6,7]. Our previous studies have investigated the effect of kaolin particle shape on the in-plane and z-direction tensile strength of coating layers [8,9] and the printing strength of papers coated with the same pigments [10]. The present study highlights the role of kaolin particle shape in determining the mechanical properties of coating layers, particularly the in-plane mechanical properties which have been shown to influence the bending stiffness and folding resistance of coated paper [11-12].

MATERIALS AND METHODS

Materials

Two pairs of coating kaolins were used having approximately similar particle size distributions as determined by sedimentation. These were chosen to cover a range of particle aspect ratio values, from blocky to very platy. The mean (d_{50}) particle size of the first set, labelled as fine, was $0.40 \mu\text{m}$ esd. Additionally, two ultrafine kaolins were included of blocky and platy morphology. These had a mean size of $0.20 \mu\text{m}$. A GCC of 95wt% < $2 \mu\text{m}$ (Carbital 95TM, $d_{50} = 0.50 \mu\text{m}$) was also used in some experiments. The properties of the kaolins are summarised in Table 1. The mean aspect ratio (defined as the average plate diameter / thickness) was measured by stopped flow conductivity using a patented technique [13]. The shape trends are generally mirrored by the difference between the average size (d_{50}) as measured by sedimentation using a Micromeritics SedigraphTM and light scattering using a Malvern MastersizerTM S. The latex binder used was a carboxylated styrene butadiene acrylonitrile copolymer of $T_g = 10^\circ\text{C}$ (DL920, Dow).

The kaolins were slurried at the optimum solids using 0.3wt% of a sodium polyacrylate dispersant (CED3546, Ondeo Nalco). The latex was added at levels between 3 and 17 pph based on clay. 0.3 pph of sodium carboxymethyl cellulose (Finnfix 10TM, CP Kelco) was also added as a thickener. After pH adjustment to 8.0, the colours were screened through $53 \mu\text{m}$. In order to remove air bubbles, the colours were centrifuged at 4000

min⁻¹ for 10 minutes. In some experiments a hydroxyethyl derivative of corn starch (Penford Gum 280, Penford Products, USA) was used in combination with latex. The starch was cooked at 20% solids at 95° - 100°C for 20 minutes.

Table I. Physical properties of kaolins

Kaolin type	Grade	Particle size distribution by Sedigraph, wt% below					d50 by light scattering (Malvern) μm	Shape factor	BET surface area, m^2g^{-1}
		2 μm	1 μm	0.5 μm	0.25 μm	d50 μm			
FB (Fine Blocky)	Blend of Capim DG / Premier	93	81	63	34	0.37	1.46	16	14
FUP (Fine Ultra Platy)	Contour 1500	90	77	60	34	0.40	3.1	56	16
UFB (Ultra Fine Blocky)	Astrasheen	100	99	94	68	0.19	0.32	10	27
UFP (Ultra Fine Platy)	Contour Xtreme	97	90	79	53	0.23	1.43	40	24

Experimental Methods

Coating films were prepared by drawdown using wire wound bars. The procedure is described in our previous publications along with the technique used for measurement of the tensile properties of the films [8,9]. Load and elongation at break were recorded for each sample, and an example of the output is shown in Figure 2. The strength was calculated by dividing the load (N) at break by the cross-sectional area of the sample across which the load is applied to give the tensile strength in force per unit area (MPa). 10 measurements were made on each sample, and the mean value calculated. In the following figures error bars signify the standard deviation. The elastic modulus of the layers was derived from the same data by taking the slope of the initial linear portion of the tensile strength / elongation plot as shown in Figure 2.

Some samples of the coatings were supercalendered using a Perkins laboratory calender. The coatings were placed between sheets of thin lens tissue to avoid the coatings sticking to the rolls. The calender was unheated and the linear load was 78 kg cm⁻¹.

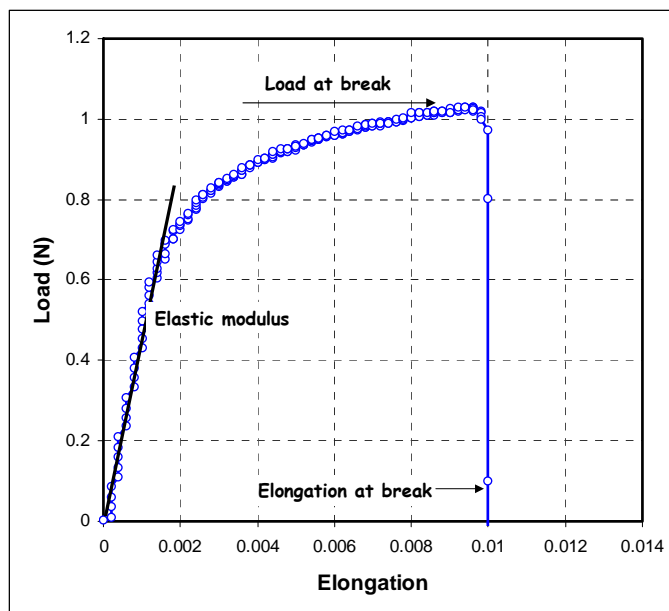


Figure 2. Example of a load / elongation curve for coating (Clay FB / 5pph latex) in the in-plane direction.

Coated papers were produced on a pilot coater running at 600 m min⁻¹. A 58 gm⁻² woodfree basepaper was used, and a coatweight of 12 gm⁻² per side applied using a roll applicator with blade metering. 50% blends by weight of each kaolin were made with a fine GCC (Carbital 95TM). 9.5 pph of styrene butadiene acrylonitrile

latex (DL920, Dow) and 1 pph CMC (Finnfix 5, CP Kelco) was added throughout. After coating, all of the coated reels were supercalendered under the same conditions using 11 nips with a surface temperature of 100°C and a linear load of 200 kN m⁻¹ at 600 m min⁻¹. The moisture of the coated papers was controlled to 5.5 ± 0.3 wt%. Coating and calendering were carried out at the pilot plant at Oy Keskuslaboratorio Centrallaboratorium AB (KCL), Helsinki, Finland.

The bending resistance of the coated papers was measured using a Stiffness Tester (Lorentzen & Wettre), following a SCAN-P test method [14]. 10 measurements of each sample were made, in the machine and cross-machine directions respectively. The results for the two directions were then averaged to give a mean bending resistance. Since the basis weights of the coated sheets was not varied, in the discussion the terms bending resistance and stiffness were taken as interchangeable.

RESULTS

The effect of particle shape on in-plane coating properties

Figure 3 shows results for in-plane tensile strength properties of coatings made from all the clays at a range of latex levels. We found that the high aspect ratio clays gave higher in-plane tensile strength than the low aspect ratio clays. The GCC coatings had the lowest in-plane tensile strength, 6-7 times lower than the strongest kaolin, the ultra high shape fine clay.

Figure 4 plots the elastic modulus of the same coatings as a function of latex level. The ranking of modulus values follows that of the tensile strength trends in Figure 3, with the high aspect ratio clays giving the highest modulus values. These high shape clays give a modulus between 5 and 8 times higher than the GCC alone.

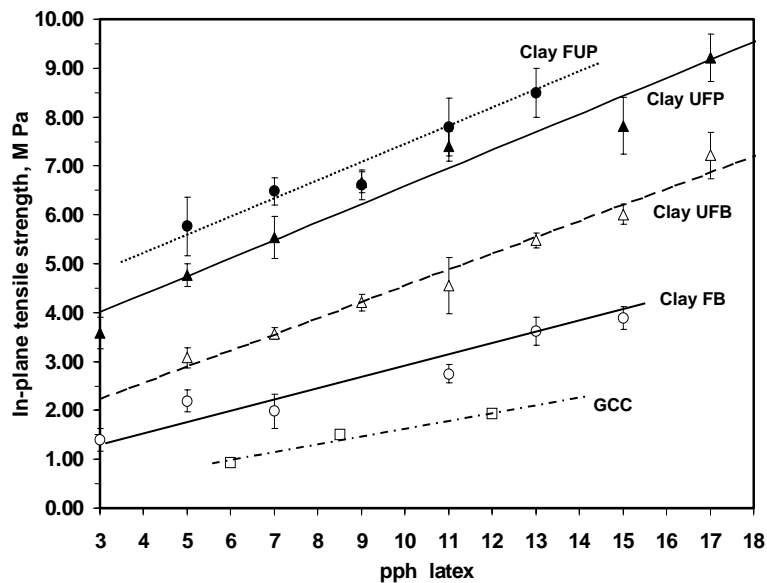


Figure 3. In-plane tensile strength of coating layers as a function of latex level.

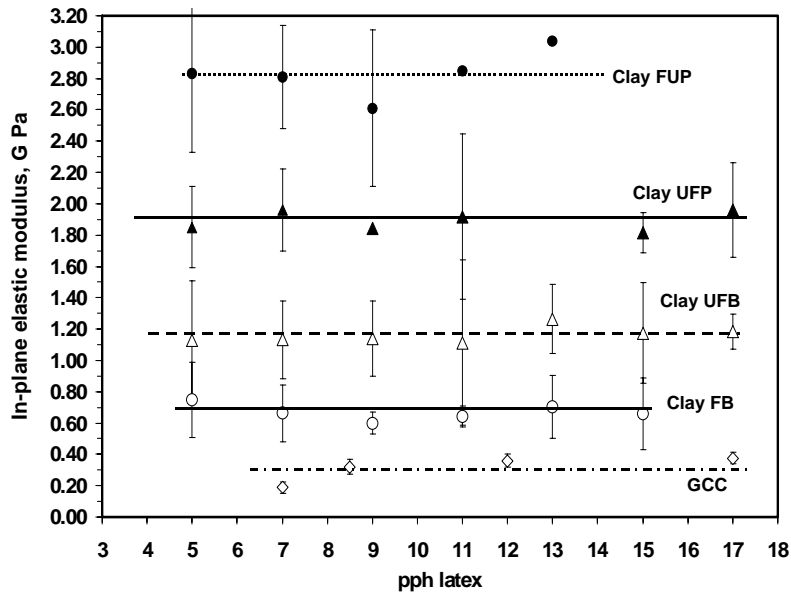


Figure 4. In-plane elastic modulus of coating layers as a function of latex level.

Calendering the coatings based on the ultrafine clays (Figure 5) showed that the in-plane strength and the modulus increased as the coating became denser. After 6 nips the coating layers reduced in caliper by 5-6 %, so for the the modulus to increase must indicate a structural change, such as increased alignment of the particles. The platy clay still held the advantage over the blocky counterpart. Also plotted in Figure 5 is the modulus of the basepaper, showing that it is unaffected by calendering, as would be expected if densification is cancelled out by caliper reduction as suggested by Kajanto [2]. Note that the final modulus values for the calendered coating layer and basepaper are both of the same order of magnitude.

It is also clear from Figures 3 and 4 that this particular latex contributes to the tensile strength but not the elastic modulus of the coatings. This is in contrast to starch (Figure 6) where we found that substitution for latex leads to an increase in modulus while lowering tensile strength slightly. These trends reflect the known influences of latex and starch on coated paper stiffness, although we stress that a high modulus latex could also be used to build stiffness [15,16].

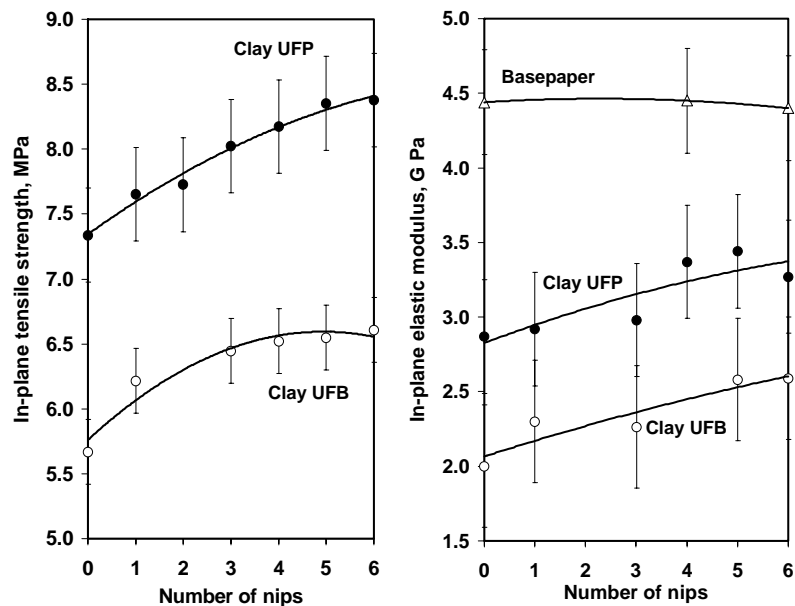


Figure 5. Effect of laboratory calendering of unsupported coating layers and basepaper on in-plane mechanical properties.

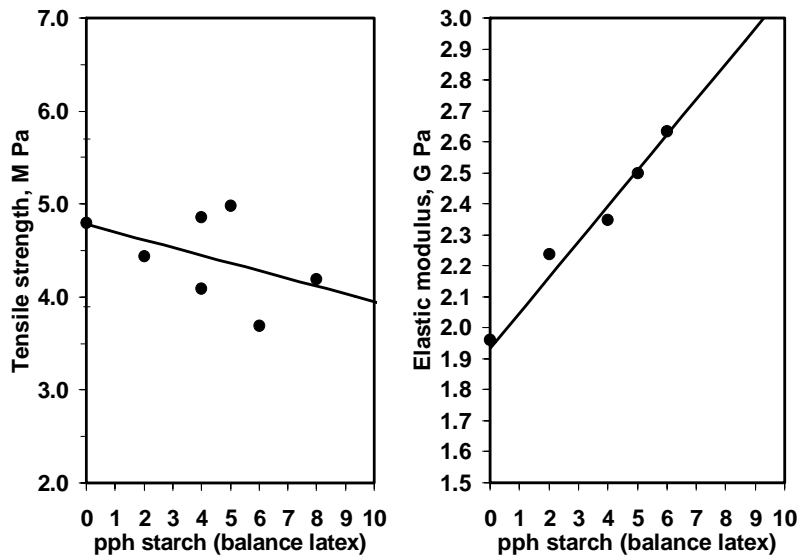


Figure 6. In-plane mechanical properties of coating layers based on UFP kaolin with a latex – starch binder system (total 10 pph).

Blends of kaolin UFP were made with a fine GCC, Figure 7. The modulus increases with kaolin addition, but the trend is not linear. These results show that kaolin can have a significant influence on the modulus even at levels of addition below 50 wt%. This is important given that in today’s formulations, high carbonate levels are required for good optical properties.

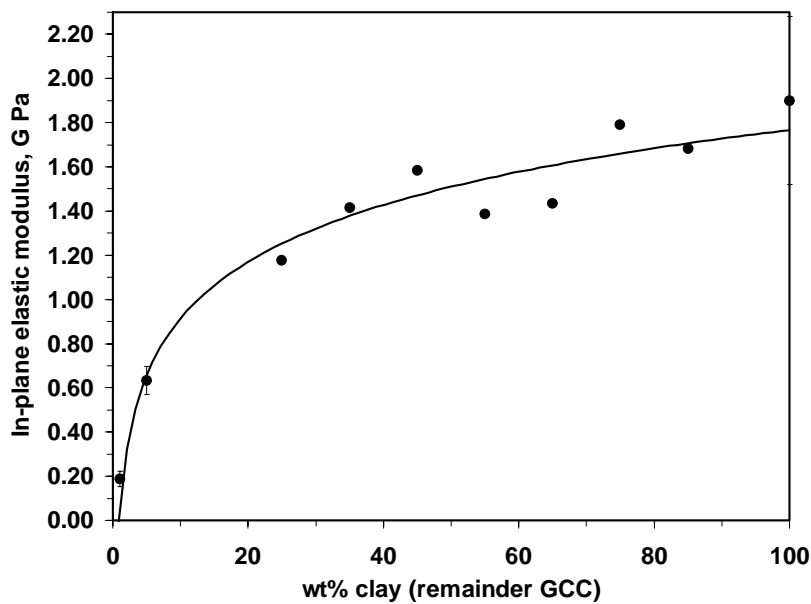


Figure 7. In-plane elastic modulus of coating layers containing kaolin and gcc of different ratios. Latex level 5 pph.

The mean bending resistance values for calendered papers coated with 50 wt% blends of kaolin and GCC from the pilot coating trial are shown in Figure 8. The trends agree with the elastic modulus values plotted in Figure 4, with the fine ultra-platy kaolin giving the greatest contribution to stiffness. We found that there was an excellent correlation between the bending resistance of coated papers containing 50% clay and the elastic modulus values for the coating layers containing 100% of the kaolin component (Figure 9).

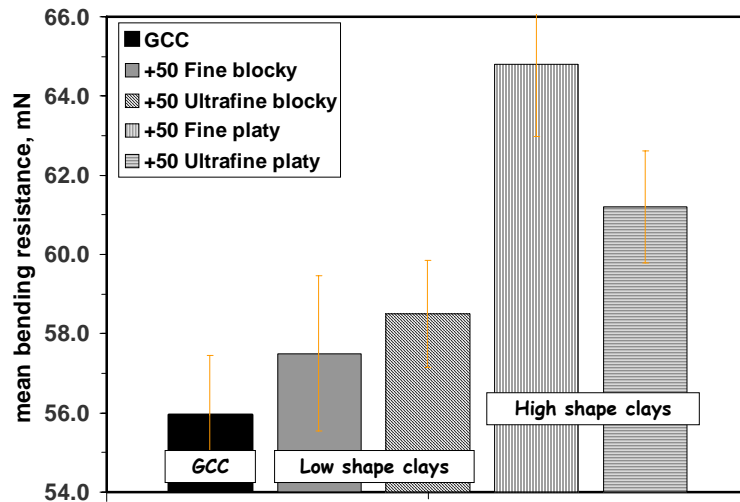


Figure 8. Mean bending resistance of coated and supercalendered papers (9.5 pph latex, 12 gm⁻² per side).

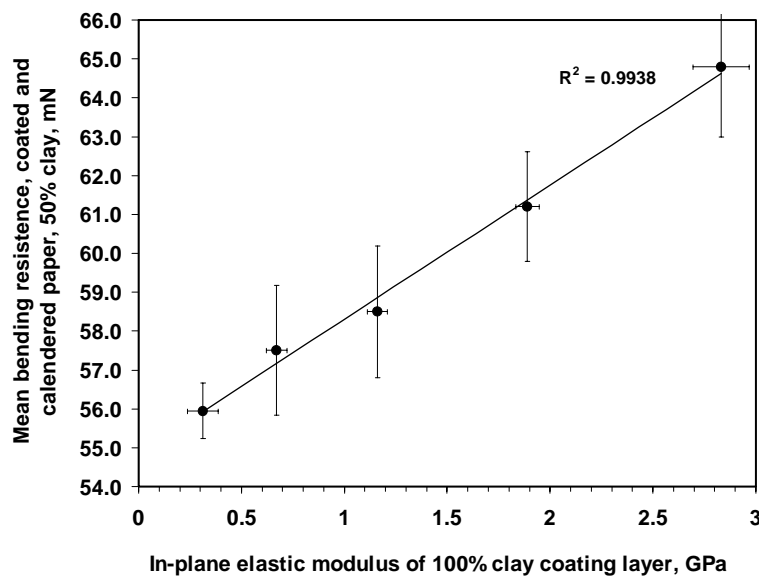


Figure 9. Correlation between mean bending resistance of coated papers and elastic modulus of coating layers.

DISCUSSION

In agreement with the published work of Lepoutre and Rigdahl [4], our study has shown that the aspect ratio of kaolin particles has a strong influence on the mechanical properties of the coating layer. Compared to widely used low aspect ratio glossing clays, high aspect ratio kaolins gave coating layer elastic modulus values which were 60 – 130% higher. The increased modulus increases the stiffness of coated papers. A pilot coater study showed that, blended 50 : 50 with GCC, the bending resistance after calendering was increased by 11 % when a high shape kaolin was substituted for a blocky glossing clay of similar particle size. Figure 10 shows the potential caliper reduction possible calculated from equation (2). The modulus value for the basepaper was fixed and the coating modulus values adjusted to give a stiffness increase of 11%. The model suggests that a 3% reduction in basesheet fibre weight would be possible.

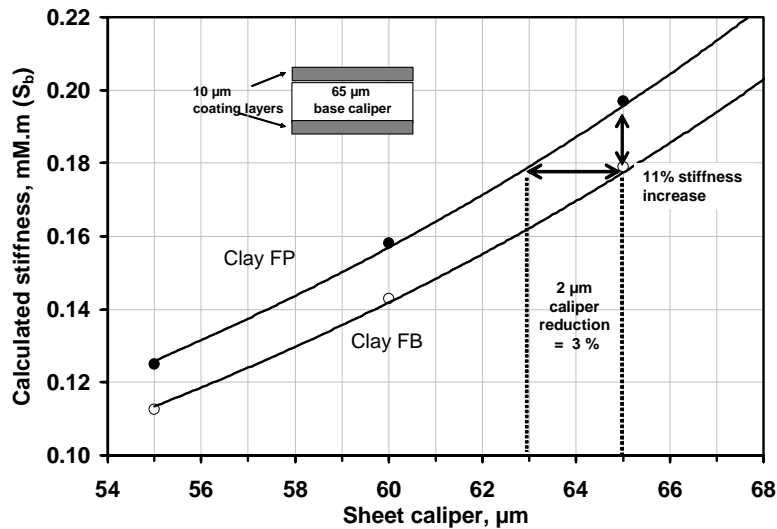


Figure 10. Calculated caliper reduction possible by improving sheet stiffness by 11%.

Recent studies by Rioux [17], Gagnon *et al.* [18], and Wygant *et al.* [19] have qualitatively confirmed these trends. This advantage of platy clays is also known from polymer science, where platy fillers are used to give enhanced flexural modulus [20,21].

An unexpected observation was that, after calendering, coating layers increased in tensile strength and modulus. This may be a result of improved particle alignment, known to occur during calendering [22]. High aspect ratio clays maintained their modulus advantage over blocky clays after calendering. The modulus of the basepaper alone did not change, suggesting that the caliper reduction keeps pace with the densification [2].

A potential drawback with high modulus coatings is that they are also more brittle and the tendency to crack during folding is increased. This may to some extent be mitigated by the increased tensile strength. There are a number of strategies that can be used to control cracking, particularly if multiple coating layers are applied [23].

CONCLUSIONS

The demands for high brightness means that calcium carbonate pigments (either ground or precipitated) are an essential component of coated printing papers today. However the approximately spherical particle shape of these mineral particles means that the stiffness contribution from the coating is quite low. We have shown that blending high shape kaolins with GCCs can give useful increases in the stiffness of coated papers. An ultraplasy clay was found to give the largest improvements in stiffness.

The role of the binder in building the elastic modulus of the coating was also investigated. Increasing the level of a standard SBR latex increased the tensile strength but had no influence on the modulus of the coating layer. Starch was found to increase the modulus of the coating. Whilst this is in agreement with the known influences of different binder systems on stiffness of coated paper, it should be emphasised that high modulus latexes which contribute to stiffness can also be used. Hence a combination of platy clay and higher modulus binders can build the modulus of the coating layer. This enables the coated paper stiffness to be increased and facilitates cost savings through reductions in the amount of fibre.

Further work should focus on the potential for fold cracking in high modulus coatings, and ways of overcoming this.

ACKNOWLEDGEMENTS

The authors thank Jon Svending and Dawn Kent for additional technical assistance, Mikael Haapanen and the crew of the KCL pilot coater, and the board of Imerys Minerals Ltd for permission to publish this work.

REFERENCES

1. J.Faust, TAPPI Papermakers and PIMA International Leadership Conference, 2007,
2. Kajanto, I, "Structural mechanics of paper and board", Chapter 6 in "Paper Physics", ed. Niskanen, K., No. 16 in Papermaking Science and Technology, Fapet Oy, Helsinki, 1998,
3. Parpillon, M., Engström, G., Petterson, I., Fineman, I., Svanson, S.E., Dellenfalk, B., and Rigdahl, M., "Mechanical properties of clay coating films containing styrene-butadiene copolymers", *J.Appl.Polym.Sci.*, **30**, 581-592 (1985),
4. Lepoutre, P., and Rigdahl, M., "Analysis of the effect of porosity and pigment shape on the stiffness of coating layers", *J.Mater.Sci.*, **24**, 2971-2974 (1989),
5. Inoue, M., and Lepoutre, P., "Influence of structure and surface chemistry on the cohesion of paper coatings", *J.Adhesion Sci. Technol.*, **6** (7) 851-857 (1992),
6. Husband, J.C., and Lyons, A.V., "Engineered coating clays for future needs", Proc. 7th Int. Conf. On New Available Technologies, 2002, pp.191-195, SPCI, Stockholm,
7. Murray, H.H., and Kogel, J.E., "Engineered clay products for the paper industry", *Applied Clay Sci.*, **29**, 199-206 (2005),
8. Husband, J.C., Preston, J.S., Gate, L.F., Storer, A, and Creaton, P., "The influence of pigment particle shape on the in-plane tensile strength properties of kaolin-based coating layers", *TAPPI J.*, **5** (12), 3-8 (2006),
9. Husband, J.C., Preston, J.S., Gate, L.F., Storer, A, and Creaton, P., "A study of in-plane and z-direction strength of coating layers with varying latex content", *TAPPI J.*, **6** (12) 10-16 (2007),
10. Husband, J.C., Preston, J.S., Gate, L.F., Blair, D., and Creaton, P., "Factors affecting the printing strength of kaolin-based paper coatings", Proc. TAPPI Coating Conf. (2007), TAPPI Press, Atlanta,
11. Kan, C.S., Kim, L.H., Lee, D.I., and Van Gilder, R.L., "Viscoelastic properties of paper coatings : structure / property relationship to end use performance", Proc. TAPPI Coating Conf. (1996), pp.49-60, TAPPI Press, Atlanta,
12. Guyot, C., Bacquet, G., and Schwob, J.M., "Folding resistance of magazine papers", Proc. TAPPI Coating Conf. (1992), pp. 255-268, TAPPI Press, Atlanta.
13. Gate, L.F., and Webb, T.W., US Patent 5,576,617 (1996),
14. "Bending resistance", SCAN-P 29:84, Scandanavian Pulp, Paper and Board Testing Committee, revised September 1984,
15. Jud, C., Kan, C., Hahn, N., "Fine tuning dynamic mechanical properties of latex and adjusting coating parameters for maximum coated paper stiffness", Proc. TAPPI Coating Conf. (2000), pp. 431-433, TAPPI Press, Atlanta,
16. Kim-Habermehl, L.H., Pollock, M., Kan, C., Oates, J., Williamson, G., "Coated paper stiffness : a practical perspective", Proc. TAPPI International Printing and Graphic Arts Conf. (2000), pp. 311-318, TAPPI Press, Atlanta,
17. Rioux, R., Bousfield, D.W., Triantafillopoulos, N., "Elastica stiffness and low load indentation measurements for the mechanical properties of coated papers", Proc. TAPPI Coating Conf. (2008), Paper 28.2, TAPPI Press, Atlanta,
18. Gagnon, R.E., Walter, J.C., Kendrick, J.W., Iyer, R.R., McLain, L., Wygant, R.W., "Metered Size Press Coating Formulation Design for Fiber Reduction," Proc. TAPPI Coating and Graphic Arts Conf. (2007) Paper 22.1, TAPPI Press, Atlanta,
19. Wygant, R.W., Kendrick, J.W. and Walter, J.C., "Metered Size Press Pigmentation for Fiber Reduction," Proc. TAPPI Coating and Graphic Arts Conf. (2008), Paper 37.1, TAPPI Press, Atlanta,
20. Adams, J.M., "Particle size and shape effects in materials science : examples from polymer and paper systems", *Clay Minerals*, **28**, 509-530 (1989),
21. Riley, A.M., Paynter, C.D., McGenity, P.M., and Adams, J.M., "Factors affecting the impact properties of mineral-filled polypropylene", *Plast. Rubber Proc. Applic.* **14**, 85-93 (1990),
22. Hiorns, A.G., Elton, N.J., Coggon, L. and Parsons, D.J., "Analysis of differences in coating structure induced through variable calendering conditions", Proc. TAPPI Coating Conf. (1998), pp.583-602, TAPPI Press, Atlanta,

23. Salminen, P., Carlsson, .R., Sandas, S., Toivakka, M., Alam, P., Roper, J., “Combined modeling and experimental studies to optimise the balance between fold crack resistance and stiffness for multilayered paper coatings, Part 1”, Proc. TAPPI Coating and Graphic Arts Conf. (2008), Paper 28.2, TAPPI Press, Atlanta.

Application statement

One of the major goals in our industry today is to reduce the amount of materials in the sheet. The most expensive components are the major targets, and to this end reducing the fibre content of paper is a key aim. Fibres give stiffness to the sheet, and this work looks at how the coating layer can be engineered to improve the overall stiffness of the coated sheet, so that fibre can be taken out or replaced by an increased loading of mineral fillers.

A recent innovation is the development of a range of high shape kaolins, and this paper shows how we can use these kaolins, which give high modulus coating layers, to increase the stiffness of coated papers. We believe they offer real value to the papermaker seeking to reduce fibre levels.



✦ **Europe**
Tel: +44 1726 818000

✦ **Asia Pacific**
Tel: +65 67 99 60 60

✦ **N. America**
Tel: +1 770 594 0660

✦ **S. America**
Tel: +55 11 2133 0055

Email
paper@imerys.com