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EVALUATION AND CONTROL OF COATED PAPER STIFFNESS

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ABSTRACT

Stiffness is an important property of paper in printing processes. Operational problems such as jamming during sheet fed offset printing often stems from poor paper stiffness. In the case of coated papers, the Young's modulus of the coating layer affects the stiffness of the whole coated paper because external layers undergo larger strain during bending compared to the internal layers.

A method to theoretically calculate the Young's modulus of a one-sided coated sheet is presented in terms of material mechanics. The accurate interpretation of Clark stiffness enables us to estimate the Young's modulus of the coating layer from Clark stiffness. The accurate calculation of Young's modulus from the Clark stiffness is shown to require an impervious film because basepaper, when wetted with water, swells and roughens, giving an incorrect value for the Young's modulus.

INTRODUCTION

Emphasis has been put mainly on printability of coated paper and fluid dynamics of coating color in the field of coating science because coating is aimed at high grade printing paper. Less research has been conducted regarding coated paper physics. However, clarifying its physical properties helps examine the bending and compression deformation that coated paper undergoes in a printing press and the durability to severe printing conditions. An example of these problems is a feeding trouble on a copy machine, optical character recognition machine and sheet-fed printing press. Namely, soft paper, especially coated paper that is likely to be with poor stiffness, congests before a destination nip.

A definite physical value, "stiffness" is used in the general analysis of materials behavior against a bend. The definition is Young's modulus E times second moment of area I, that is, EI, is the stiffness of a material against a bend. Then, when one says that this paper is weak, this "weak" or "strong" is not definitive, and just subjectively sensed by people. Though this kind of "weak" or "strong"("sensory stiffness" refers to this concept thereafter) is considered to be correlated with stiffness, Kazumori et. Al[1)]reported that at the same stiffness EI, the higher the Young's modulus E, the "harder" the paper was sensed. They added, however, that examinees evidenced that the criteria of sensory stiffness is resistance to a snap and slow flip both due to the weight of its own, thickness and recovery property from a bend as well as the resistance to an external bending force.

Physical values related to stiffness are summarized in the following manner. "Clark stiffness" means a bending resistance in the case that a paper strip bents due to the weight of its own. "Pure bending stiffness"[2)3)] means a stiffness divided by the sample width in the case that a paper strip is bent with the same curvature along the whole span (The closer to the support, the higher the curvature in Clark stiffness.). "Liveliness" originally indicates the speed at which a textile recovers from certain curvature. Naito et al. [4] applied this to paper and defined the liveliness of paper to be "an inverse of recovery time squared" so that this immediate recovery property would be connected with stiffness. Liveliness and Clark stiffness may be more suitable to analyze paper behavior on a off-set press feeding sheets with suction pads because how fast the paper sheet recovers after it bents due to the weight of its own and the moment of inertia of the paper is relatively important. **Table 1** lists some physical values regarding stiffness including those mentioned above.

Table 1. Terminology related to stiffness

Physical value	Equation	Concept
Stiffness	S, EI	Resistance to a bend
Clark stiffness (Handling stiffness)	$\frac{L^3}{100}, \propto \frac{EI}{bW}, \frac{ET^3}{12W}$	Resistance to a bend due to the weight of its own
Flexing resistance	$\frac{L^3W}{100}, \propto \frac{EI}{b}$	Stiffness divided by sample width
Pure bending stiffness	$\frac{EI}{b}$	Stiffness divided by sample width when bent with the same curvature along the whole span
Liveliness	$\frac{1}{t^2}$	Inverse of squared time of recovery from certain curvature
Sensory stiffness		Stiffness subjectively estimated by people
Young's modulus	E, $\frac{\sigma}{\varepsilon}$	longitudinal elastic modulus
Second moment of area	I, $\frac{bT^3}{12}$	Integral of squared distance from neutral axis
Bending moment	M, $\frac{EI}{r}$	Force times distance

where, L is overhung length (cm); W basis weight (g/m^2); t time (s). T is thickness; b is sample width; σ is stress; ε strain; r radius of curvature.

There has been a lot of research regarding stiffness of uncoated paper, but few regarding that of coated paper. Naito et al.[5] measured stiffness of coated paper in several methods and compared them as well as uncoated paper, then concluded that pure bending stiffness is more correlated to sensory stiffness than Clark stiffness. Nagai et al. [6] calculated the Young's modulus of the coating layer of two-sided coated paper from Gurley stiffness, but it can not be applied to one-sided coated paper which are regularly produced on a laboratory scale.

Basepaper property dominates most mechanical properties of the whole coated paper. However, particularly with regard to stiffness in a bending mode, a coating layer has considerable effects [7][8]. This is because the strain the outlying coated layer undergoes is much larger than that the inner basepaper layer does. However, because the relative density of dry coating is about twice as large as that of basepaper, Clark stiffness decreases with increasing coat weight even if Young's modulus of the two layers were equal.

Considering that coated paper a composite material consisting of the two layers, that is, coating and basepaper, this work aims at the estimation of how much the coating layer contributes to the total stiffness of the coated paper. For this purpose, we assumed that determination of Young's modulus of a coating layer would be the best approach, because it is independent of the coat weight and thickness, but is dependent only on the color formulation and coating structure. In the beginning, theoretical and empirical methods were established to determine Young's modulus of a coating layer. Then, effects of starch and plastic pigment formulation on Young's modulus of the coating layer were examined. Finally, double coating with colors having different color formulations was evaluated in view of improvement of stiffness of the coated paper.

THEORY

It is assumed that coated paper is a composite material having two parallel layers of uniform and homogeneous substance glued together. Nagai et al.[6] showed the equation to calculate Young's modulus of the coating layers for two-sided coated paper, assuming the same coat weight on the two sides. However, it is usually difficult to coat a sheet to meet this requirement on a laboratory scale. Therefore, another equation for one-sided coated paper for it was led as mentioned below.

The total stiffness R_a is the average Young's modulus E_a times second moment of area I_a of the whole coated paper (See **Figure 1**.).

$$R_a = E_a \cdot I_a = E_a \frac{bT^3}{12} \quad , \tag{1}$$

where T and b are coated paper thickness and width, respectively.

Second moment of area of the coated layer I_c, its Young's modulus E_c, second moment of area of the basepaper layer I_f *and* its Young's modulus E_f give the equation (2).

$$R_a = E_c \cdot I_c + E_f \cdot I_f \tag{2}$$

Then, the reference axis (y-axis) is taken at the bottom of the cross section to determine the coordinate of the neutral axis N-N, where no strain occurs. The bending stress develops only in z-axis and the integration of the stress with regard to y_0-axis (y-N) is 0. Consequently, equation (3) is obtained.

$$E_f \int_f y_0 dA_f + E_c \int_c y_0 dA_c = 0 \quad , \tag{3}$$

where A_f and A_c are area of the basepaper and the coating layer, respectively. Substituting $y_0=y-N$ into equation (3) leads equation (4).

$$E_f \int_f (y-N) dA_f + E_c \int_c (y-N) dA_c = 0 \tag{4}$$

If this is solved with regard to *N*, Equation (5) is obtained.

$$N = \frac{E_f \int_f y dA_f + E_c \int_c y dA}{E_f \int_f dA_f + E_c \int_c dA_c} = \frac{E_f \int_0^{h_f} ybdy + E_c \int_{h_f}^{h_f+h_c} ybdy}{E_f bh_f + E_c bh_c}$$

$$= \frac{E_f h_f{}^2 + E_c \left(2h_f h_c + h_c{}^2\right)}{2E_f h_f + 2E_c h_c} \quad , \tag{5}$$

where h_f and h_c are thickness of the basepaper and coating layer, respectively. Equation (2) and (5) give Young's modulus of the coating layer E_c as shown in equation (6). Therefore, E_c can be calculated if one measures stiffness of coated paper R_a.

$$E_c = \frac{R_a - E_f \int_{-N}^{h_f - N} y^2 dy}{\int_{h_f - N}^{h_f + h_c - N} y^2 dy} = \frac{3R_a - E_f (h_f - N)^3 - E_f N^3}{(h_f + h_c - N)^3 - (h_f - N)^3} \qquad (6)$$

Clark stiffness has been known as a standard method to measure stiffness of paper or textile for a long period. Following is how to calculate Young's modulus of a coating layer from it. First, Clark stiffness is defined this way. A long paper strip is nipped at one end and supported upright. As the nip rotates, the strip flops suddenly from the bend side to another at certain nip angle. The angles where this occurs on both the sides are measured. The overhung part is extended until the difference of the angles reached 90 degrees. The satisfactory overhung length *L* cubed divided by 100 is a Clark stiffness. In Clark's publication[9)] and JIS(Japan Industrial Standard) [10)], *L* is measured in cm, but in mm in Tappi test methods[11)]. The reason why the cubed length is used is expressed in equation (7), as Oda et. al[12)] showed.

$$\frac{L^3}{100} \propto \frac{EI}{bW} \quad \left(= \frac{ET^3}{12W} \right) \qquad (7)$$

According to their work, the overhung length *L*(cm), Young's modulus *E*(dyne/cm^2), thickness *T*(cm), basis weight *W*(g/m^2) empirically satisfies the equation where the two terms are equal (not only proportional) in the condition that those variables are expressed in the specified units above.

Theoretically, as Takadera et al. [13)] showed, this action is represented by an equation of a beam with one end fixed, equation (8),

$$\frac{d^2\theta}{dq^2} = q \sin\theta \qquad (8)$$

(Boundary condition: q=0 at θ=α and

$$q = K = \sqrt[3]{\frac{b(W \times 10^{-4})G}{EI}} L \quad \text{at } \theta = \beta) \qquad (9)$$

where, q, θ, α, β is a distance from the free end, an angle from the downward vertical direction at q, an angle at the free end and an angle of the nip at the fixed end, respectively. If the nip angle is even on the right and left sides, β is equal to 225 degrees. All the other variables are equal to that of equation (7). They calculated that a paper strip flops at this angle at a condition, K=2.71. K is correspondent to a kind of corrected overhung length which has no unit and is independent of Young's modulus and thickness of the material. Practically, since basis weight acts as a load to bend a paper strip, it must be multiplied by the gravitational constant G. Thus, equation (10) is obtained regarding Clark stiffness by substituting K=2.71 into equation (9).

$$\frac{L^3}{2.71^3} = \frac{ET^3}{12G(W \times 10^{-4})}, \quad \frac{L^3}{203} = \frac{EI}{bW} = \frac{ET^3}{12W} \qquad (10)$$

In the first equation of equation (10), every unit is equal to that of equation (7). To adjust every variable to the cgs system of units, basis weight was corrected as $W \times 10^{-4}$(g/cm^2). In addition, G=981(cm/s^2). In the second equation, the units, basis weight in g/m^2, length and thickness in cm and Young's modulus in dyne/cm^2, can be used. Actually, draft and the vibration of the paper strip may prompt it to flop earlier than it would otherwise. Thus, a little shorter *L* may be measured than the true *L*.

Leading a general empirical equation is very difficult because the coefficient varies depending on the method in which Young's modulus and thickness are measured, as Oda et al. discussed. In our preliminary experiment to check if this equation is right, polyethylenetelephthalate (PET) film having uniform thickness was subjected to Young's modulus calculation because thickness of paper varies depending on the measurement method. **Table 2** shows that Young's modulus calculated based on theoretical equation (10) came close to the dynamic Young's modulus measured by the ultrasound velocity method, but is about twice as large as that in the reference. In contrast, Young's modulus calculated based on the empirical equation (both the sides assumed to be equal in equation (7)) was considerably larger than the dynamic Young's modulus. In this work, equation (10) is used $R_a(=EI)$. Then, equations (4) and (5) are used to calculate Young's modulus of a coating layer.

Table 2. Young's Modulus of Polyethylenetelephthalate (PET) film

Method and equation	Clark $\frac{L^3}{100}=\frac{EI}{bW}$	Clark $\frac{L^3}{203}=\frac{EI}{bW}$	Ultrasound velocity	Reference* (Bending mode at 23 ℃)
Modulus, GPa	12.8	6.3	7.2	2.41～3.10

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EXPERIMENTAL

Table 3 shows the coating color formulation used. Calcium carbonate (Brilliant-15, Shiraishi Industry), Starch (Ace-A, Oji cornstarch), SB-latex (TO-135, Mitsui Toatu Chemicals) and Dispersant (Aron T-40, Toa Gosei Chemicals) were used. Basestock was commercial wood-free fine paper (25 s Stöckigt sizing degree and 64 g/m^2 basis weight) and impervious polyethylenetelephthalate (PET) film 100 μm thick to apply coating uniformly. Hereafter this basepaper and PET film will be referred-to as just basepaper and basefilm, respectively. Coating was applied manually with wire bars, #6, 10 and 16, Kumagai Riki Kogyo, then dried at an atmospheric temperature. Those were conditioned at 20 °C and 65 % relative humidity for more than 24 hours (Hereafter 'conditioning' refers to this condition.) and subjected to measurements.

Table 3. Formulation of coating color

Constituent	parts
Calcium carbonate	100
SB-latex	10
Starch	5
Sodium polyacrylate	0.8

Solids content = 50%

In the experiment to examine effects of water absorption to the basepaper, distilled water was applied with a wire bar #10 to about 10 g/m^2, then dried at an atmospheric temperature. Pure Bending stiffness was measured after samples were conditioned.

Coated paper and coated film were cut into a 5 mm×5 mm square with one edge cut sharply at 45 degrees to be observed. The specimen was adhered on a specimen stub for SEM with two-sided conductive tape, then platinum-coated for 300 s in an ion spatter, E-1030, Hitachi and observed on a field emission type microscope, S-4000, Hitachi.

To prepare a coated film with rough coating/film interface, basefilm was hot-pressed at 200 °C for one minute

together with a sandpaper sheet, Teflon board, iron board sandwiched in this order outward on either side, as illustrated in **Figure 2**. The basefilm hot-pressed without a sandpaper sheet was compared to it because the hot-press changed Young's modulus of basefilm a little. These basefilm sheets were coated with the color of Table 3 and dried at an atmospheric temperature.
Clark stiffness was measured according to JIS P 8143. The sample width was 30 mm. The results were rounded to three orders of a significant figure.

Pure bending tester has a characteristic that a paper strip is bent with the same curvature along the whole span. This tester measures early stiffness at the beginning of a bend and average stiffness. In this work, only early stiffness is plotted in every figure. Coated products were cut basically to a 100 mm×100 mm square after conditioning. The sample was set on a tester with a span of 50 mm in Machine direction in the case of paper and a width of 100 mm. The tester used was pure bending tester JTC-1, Nihon Seiki.

Young's modulus of a coating layer was calculated according to equations (5) and (6) from Clark stiffness and according to the equation, $S_{PB} = ET^3/12$ from pure bending stiffness. Young's modulus of a coating layer was examined by changing colors formulation.

Binder mixture was prepared only with starch and SB-latex. The mixture ratio was five sorts; starch:SB-latex = 0:15, 3:12, 5:10, 7:8 and 10:5. Those were applied on basefilm, then dried at an atmospheric temperature. Clark stiffness of the coated film was measured after conditioning.

Calcium carbonate of the color of Table 3 was all replaced with plastic pigment(PP) here. The PP used was polystyrene emulsion P-2 and P-4, Mitsui Chemicals, with characteristics listed in **Table 4**. The color was prepared to be 27 to 33 % solids and applied on basefilm then dried at an atmospheric temperature. Clark stiffness was measured after conditioning.

Table 4. Resin constants of plastic pigments

Plastic pigment	P-2	P-4
Appearance	Milky-white	Milky-white
Major Ingredient	Styrene	Styrene
Structure	Dense (not hollow)	Dense(not hollow)
Involatile ingredients, %	47.9	46.9
PH	8.0	8.1
Viscosity, cps	41	15
Particle diameter, μm	0.23	0.50

Basefilm was used to make uniform coatings. Two kinds of colors shown in **Table 5** were used. First, color SF was applied manually with a wire bar #16 and dried at an atmospheric temperature. Then, color SC was coated additionally on it in the same way after the first layer was well dried. Another kind of sample coated in the reversed order of the two colors was also prepared. Pure bending stiffness was measured.

Table 5. Formulation of coating color for double coating

Color type	SF(Starch-free)	SC(Starch-containing)
Calcium carbonate	100	100
SB-latex	15	10
Starch	0	5
Sodium polyacrylate	0.8	0.8

unit: parts
Solids content = 50%

To estimate a general range of Young’s modulus of basepaper, several wood-free and wood-containing basepapers were used.

RESULTS AND DISCUSSION

Figure 3 shows Clark stiffness and pure bending stiffness of the coated film and pure bending stiffness of the coated paper. Clark stiffness has a dimension of stiffness divided by the weight of its own. Consequently, it decreases as coat weight increases because Young's modulus of the coating layer is low for its high relative density. The data scattered because it is difficult to apply coating uniformly by hand and the results depended on whether the thick side was nipped or the thin side. Pure bending stiffness increased additionally by the amount of stiffness that the coating layer shared with increasing coat weight.

Figure 4 shows Young's modulus of the coating layer calculated from the three kinds of stiffness in Figure 3. Young's modulus of the coating layer on the film was about 0.7 GPa from Clark stiffness and 0.5 GPa from pure bending stiffness. These data scattered to some extent for the same reason as before - nonuniform thickness, but did not depend on the coat weight.

On the other hand, the coat weight dependence was found with the coating on paper. Some of them resulted in negative values below about 17 g/m^2 coat weight. This apparently stems from that Young's modulus was miscalculated. The possible reasons for this are: (1) increased thickness and decreased Young's modulus due to water absorption, (2) moisture content change even in the same atmosphere due to water absorption, (3) Pore-filling near basepaper surface with a coating and a roughened coating/basepaper interface due to dehydration from a wet coating, (4) Changes in coating composition and structures during consolidation processes and the difference in Young's modulus from a coating on film. If reason (4) is true, this is special to paper and the evaluation of a coating on film is not applicable to the paper case. However, reasons (1) to (3) are true, coating on paper is not appropriate to evaluation of Young's modulus of a coating layer. From here, those possible reasons were examined.

Table 6 shows changes in physical properties of basepaper that absorbed distilled water. Water absorption swelled the basepaper by 6 % in thickness. The basis weight was increased 2 % because of the hysteresis via high moisture content (wetting). Then, Young's modulus was reduced by 34 %. This suggests that Young's modulus of unwetted basepaper cannot be used as an alternative Young's modulus of the basepaper layer, thus miscalculating Young's modulus.

Table 6. Changes in sheet properties of wood-free paper by water application

	No water appl.	After water appl.
	Average (Std. dev.)	Average (Std. dev.)
Thickness, μm	80.2 (0.5)	85.0 (0.8)
Basis weight, g/m^2	64.3 (0.6)	65.6 (0.5)
Pure bending stiffness, gf・cm	1.22 (0.06)	0.95 (0.13)
Young's Modulus, Gpa	2.77 (0.12)	1.82 (0.24)

Figure 5 shows pure bending stiffness of the coated film with a rough or smooth coating/film interface. Stiffness of the coated film with a smooth interface was larger than that for a rough interface. This means that the smoother the interface, the larger the coated sheet stiffness, thus implying that Young's modulus of a coating layer would be miscalculated if the coating was applied on basepaper due to roughening[14)].

Scanning Electron Micrographs, **Picture 1 and 2**, are cross sections of the coated paper and the coated film, respectively. It is revealed that the coating filled the surface pores of the basepaper and that the interface is rough, while the coating/film interface is very smooth and the thickness is constant. This exemplified that this difference was one of the reasons why the calculated results of Young's modulus were different between paper and film in Figure 4.

Figure 6 shows Young's modulus of the blend film made on basefilm only from starch and SB-latex. With increased starch ratio, the blend film Young's modulus increased remarkably. The ratio 33 % corresponds to the binder composition of the color used in Figure 4. The Young's modulus of the blend film then was 0.60 GPa, being close to the average of Young's modulus (0.66 GPa) of the coating layer of Figure 4.

Figure 7 shows Young's modulus of the coating layer containing only plastic pigment(PP) in place of calcium carbonate as a function of formulated parts of starch (The total binder parts was 15). Remarkable starch effects on Young’s modulus were found with the two PPs irrespective of the particle diameter (0.23 μm and 0.50 μm). Some data of Young's modulus was calculated as negative values, presumably because the nonuniform coating (thick parts locally present) misled the thickness to a larger value than the average. The larger the particle diameter of PP, the higher the Young's modulus. It is probably because the small particle made the structure tighter during consolidation. Young's modulus of the coating containing only calcium carbonate as pigment is also plotted from Figure 4. Young's modulus of the coating in the case of the smaller PP was close to that in the case of calcium carbonate. However, PP is expected to make Clark stiffness (stiffness against the weight of its own) of the whole coated paper larger than mineral pigments even if Young's modulus of the coating layer is on the similar level because PP is lighter.

Figure 8 shows Young’s modulus of basepaper. Ash contents of wood-free A(WF-A) is higher than wood-free B(WF-B). Generally, Young’s modulus of base paper are between 0.5 and 3.5 GPa and depends on pulp formulation, pulp freeness, fiber orientation, size press and so on. As shown on before, the case of coating layer, Young’s modulus is between 0.1 and 2.0 GPa depends on binder formulation, binder contents, pigment packing and so on.

As a measures to improve coated paper stiffness, external coating of high Young's modulus containing a higher ratio of starch was tried. **Figure 9** shows pure bending stiffness of the coated film versus the total coat weight. The coated film with the starch-rich (SR) coating external exhibited higher stiffness than internal. This suggests that external coating of a color containing constituents to be stiff acts to improve the stiffness of the whole coated paper. Practically, too much formulation of starch to the external coating should reduce the pick strength. Consequently, care must be taken in deciding the right formulation ratio of starch to maintain other properties.

CONCLUSIONS

1. The equation was led to calculate Young's modulus of a coating layer from stiffness and thickness both of a one-sided coated sheet and only basesheet (paper or film). Then, Clark stiffness was interpreted theoretically and accurately, then the method by which Young's modulus of a coating layer was calculated from Clark stiffness was established.
2. Young's modulus of the coating layer on *paper* was found to be miscalculated. It is because thickness (due to swelling) and basis weight (due to hysteresis) increased after coating and those values of unwetted basepaper were not applicable to calculation. Therefore, impervious film must be used.
3. Young's modulus of the blend film made from starch and SB-latex increased with increased starch ratio. Starch helps improve Young's modulus of a coating layer. Plastic pigment of the smaller particle diameter made the coating Young's modulus higher, but was close to that of calcium carbonate. However, light PP could improve stiffness against the weight of its own.
4. External deposit of starch-rich (stiff) coating improved stiffness of the whole coated film.
5. Young’s modulus of base paper is between 0.5 and 3.5 GPa and depends on pulp formulation, pulp freeness, fiber orientation, size press and so on. The case of coating layer, Young’s modulus is between 0.1 and 2.0 GPa depends on binder formulation, binder contents, pigment packing and so on.
6. Particularly with regard to stiffness in a bending mode, a coating layer has considerable effects. This is because the strain the outlying coated layer undergoes is much larger than that the inner basepaper layer does.

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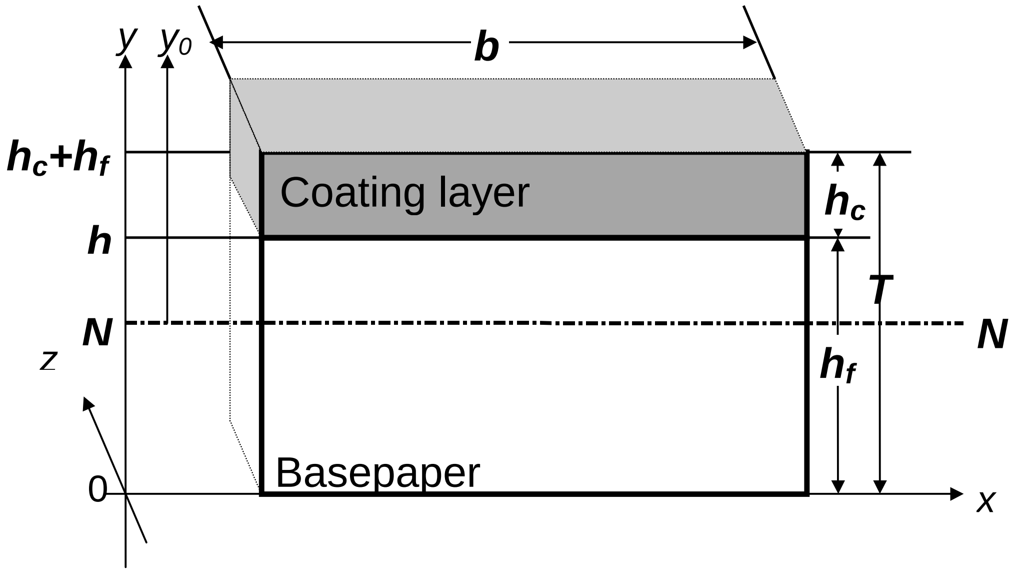


Fig. 1. Schematic cross section of coated paper for calculating second moment of area.

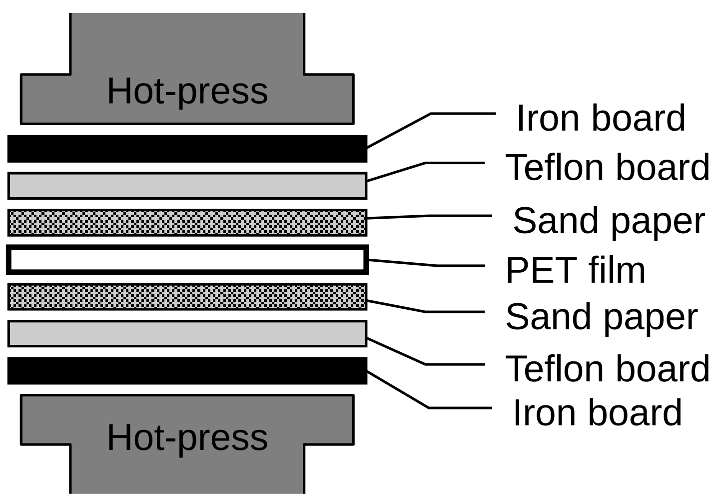


Fig. 2. Order of sheets in hot-press to make a rough

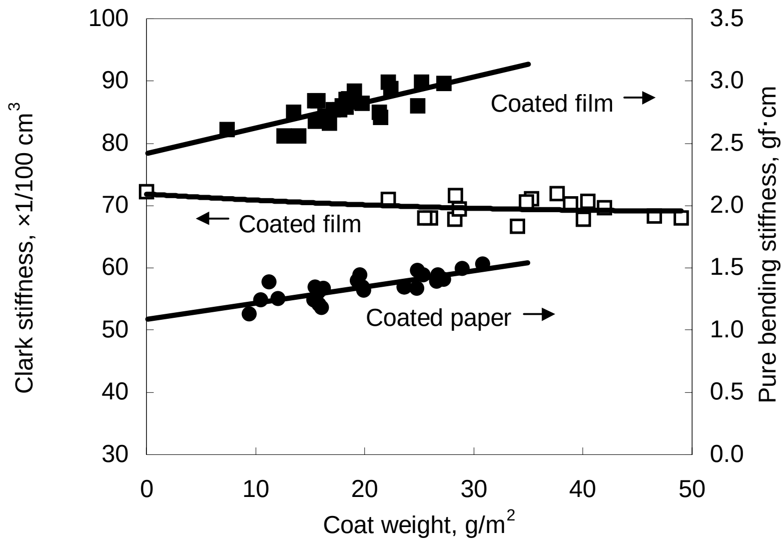


Fig. 3 Clark stiffness and pure-bending stiffness of coated paper and film as a function of coat weight.

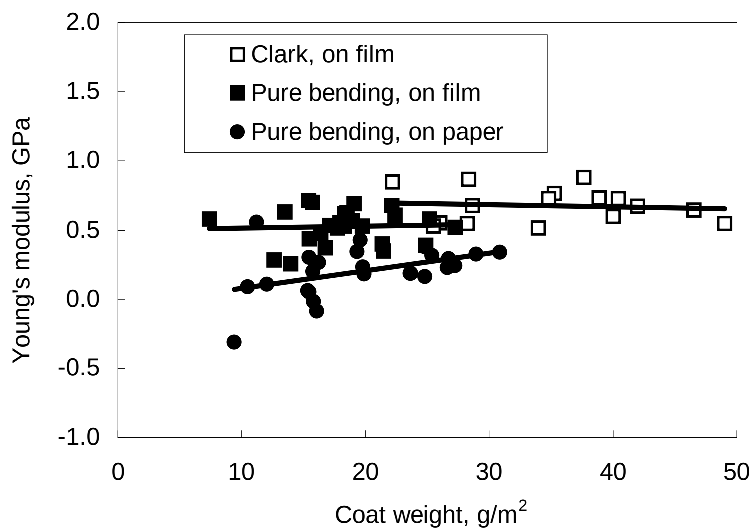


Fig. 4 Calculated Young's modulus of coating on paper or film as a function of coat weight.

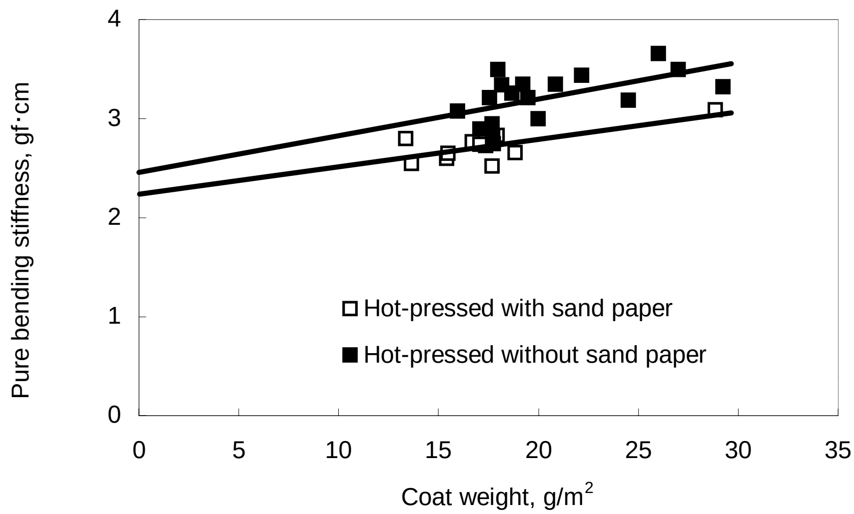


Fig. 5 Influence of interface shape on relationship between basis weight of coated film and pure bending stiffness.

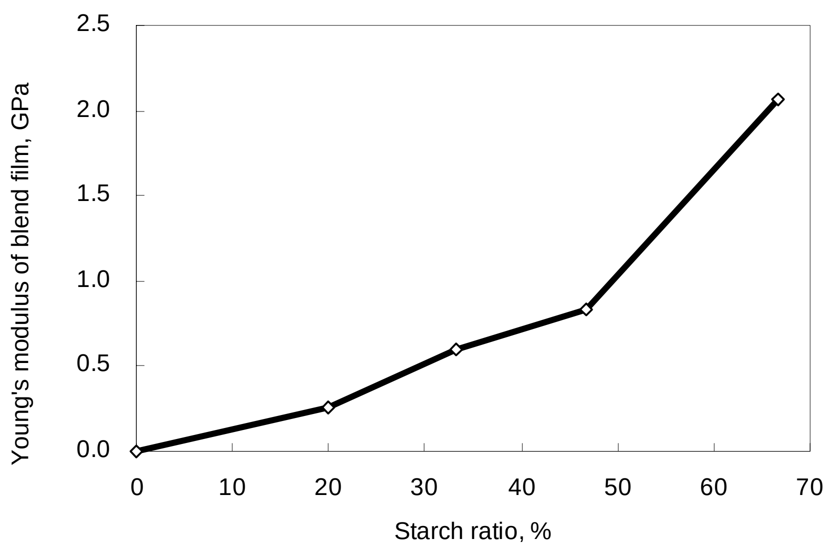


Fig. 6 Stach effect on modulus of latex/starch blend film

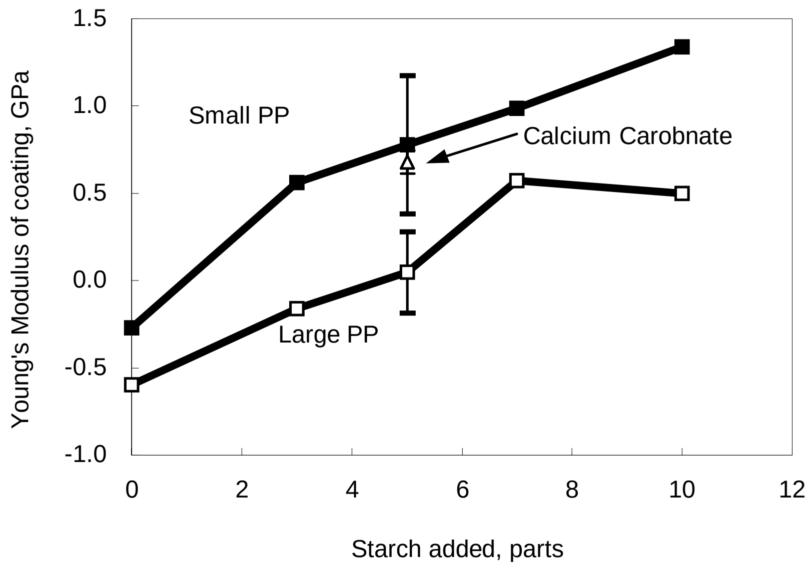


Fig. 7 Starch and plastic pigment(PP) effect on coating modulus.

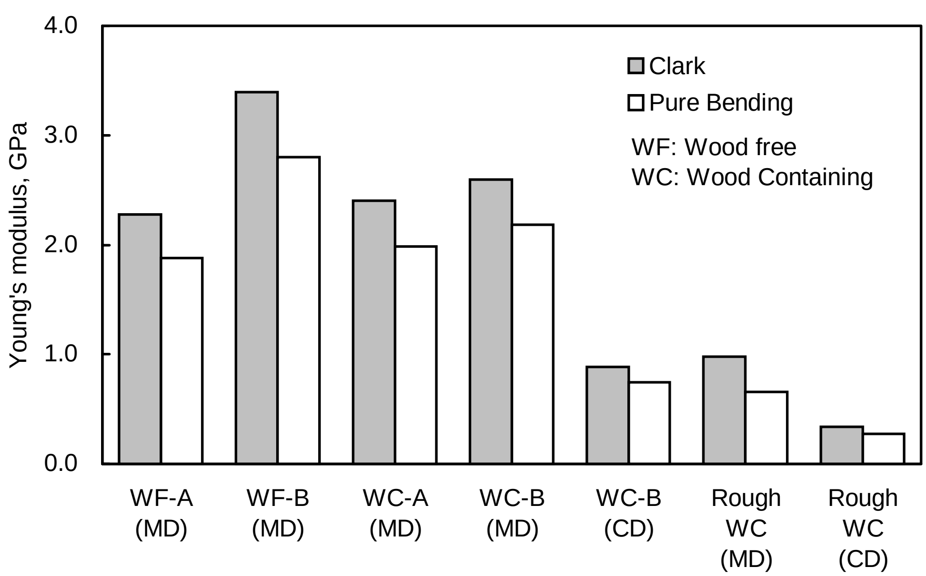


Fig.8 Young's Modulus of Basepaper

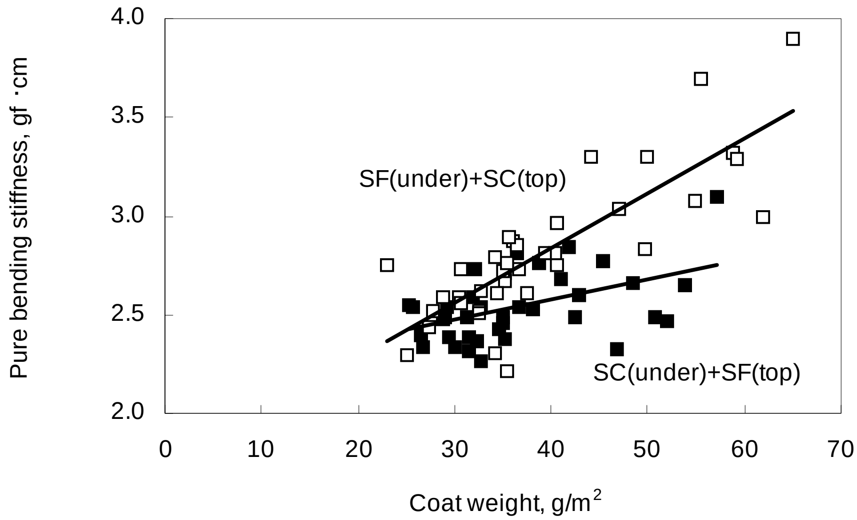
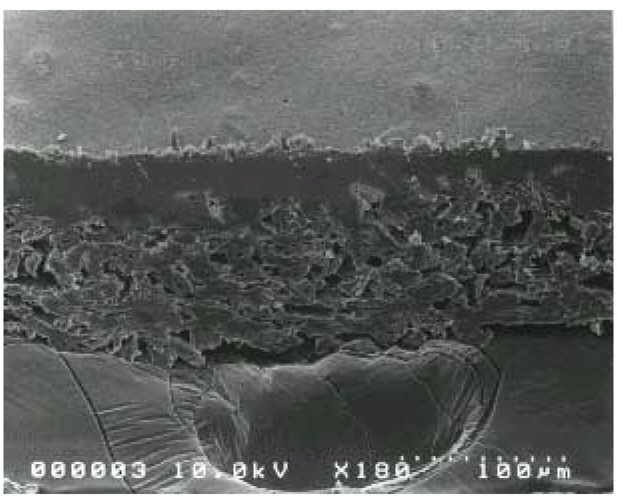
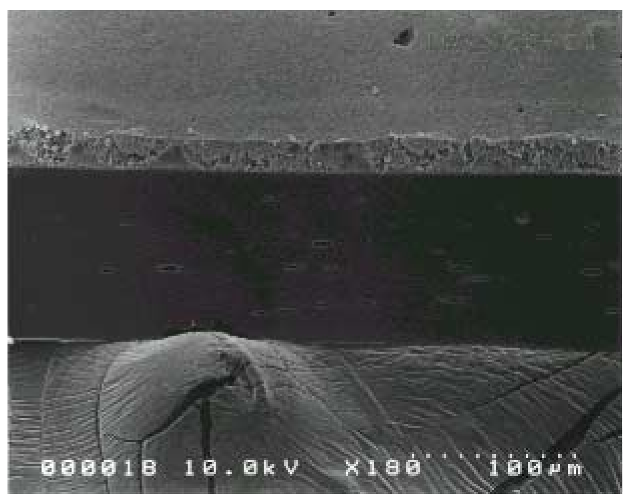


Fig.9 Effect of double-coating order of starch-containing(SC) and starch-free(SF) colors on pure bending stiffness of coated film.



Picture 1. Cross section of coated paper



Picture 2. Cross section of coated film.