

Micro-model of paper

Part 3: Mosaic model

Wentao Lu, Leif A. Carlsson, and Alf de Ruvo

ABSTRACT: *In this third and final part of the series, a simplified mosaic model is introduced for predicting paper properties. Model predictions are compared with experimental data for papers made from bleached chemical pulp, groundwood pulp, and linerboard. The experimental results generally fall within predicted bounds and are in reasonable agreement with predictions from the mosaic model.*

KEYWORDS: *Bleached pulps, chemical pulps, composites, formation, ground wood, liner boards, mathematical models, microstructure, moisture content, paper properties, paper sheets, paper structure, simulation.*

In Part 1, we derived elastic constants and moisture expansion coefficients of assemblages of cell-wall material configured in series and in parallel (1). In Part 2, microstructural parameters of paper such as volume fractions of bonded material and free fiber segments were calculated using Monte Carlo simulation of sheet formation and structure (2). Our objectives in Part 3 are (a) to combine the information gathered in the previous parts into a mosaic model that contains bonds, free segments, and voids and (b) to predict the response of paper from this model.

The “mosaic,” or “aggregate,” model was initially proposed by McGee and McCullough for model-

ing short-fiber composites (3). McCullough pointed out that the mechanical behavior of short-fiber composites (that resemble paper) can be predicted only if a complete description of the microstructure is available (4). Because such information rarely exists, micro models for complex materials should be based on structural descriptors that can be evaluated experimentally. The complex and variable features of the microstructure of short-fiber composite materials led to the development of the aggregate model, in which the material is viewed as an assemblage of constituent materials.

As discussed in Part 1, previous network and continuum models of paper are based on the assumption

that the strain field is uniform throughout the entire sheet. In reality, however, stiff regions will deform less than average strain field, and compliant regions will undergo more deformation than the average strain field. Recently developed computer methods for enhancing vision have allowed researchers to detect substantial variations in local strain in tension test specimens (5).

To overcome some of these difficulties, we present a mosaic model of paper. The mosaic model is used for predicting the elastic and hygroexpansion properties of paper. Model predictions are compared with experimental data for papers made from chemical and groundwood pulps and linerboard.

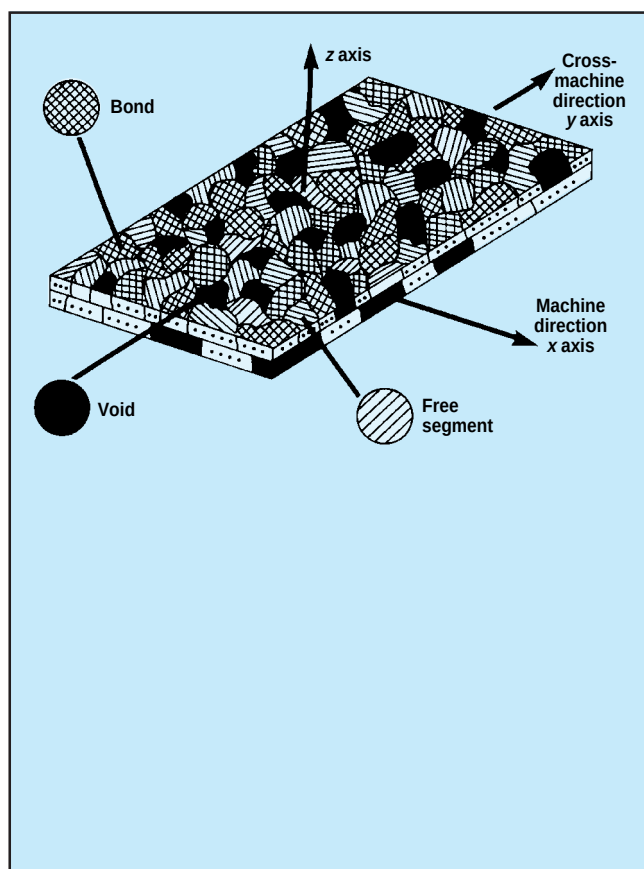
Mosaic representation

The microstructure of paper is represented by the mosaic model shown in **Fig. 1**. The *x* axis represents the machine direction (MD), and the *y* axis represents the cross-machine direction (CD). The *z* axis is the thickness coordinate. The three “phases” that compose the model are fiber crossings, fiber segments, and voids. A free fiber segment is the unbonded length of the fiber between the bonded regions. A fiber crossing, or a bonded region, is the area where two fibers are attached to one another.

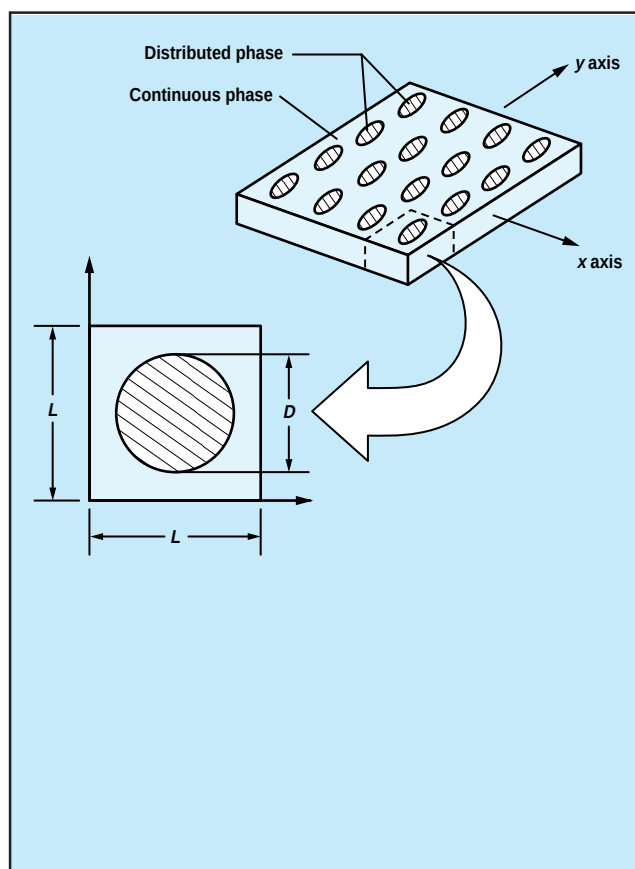
The fiber crossings and segments are both composed of cell-wall layers. The S_2 layer is assumed to dominate the response of the cell wall. As

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1. The structure of paper in the mosaic concept



2. The simplified representation of paper structure in the mosaic concept, with a blowup of a representative volume element



discussed in Part 1, the S_2 layer is treated as a composite of unidirectional microfibrils in a matrix of hemicellulose and lignin. The fiber crossings are considered as elements of cell-wall material bonded together as multi-directional laminates, thus being connected in a parallel loading arrangement. Such a configuration leads to upper bounds on the elastic moduli (1). The free segments are considered as cell-wall elements arranged in series, which leads to lower bounds on the elastic moduli (1).

In Part 1, the elastic constants and moisture expansion coefficients for such assemblages of cell-wall elements were derived as functions of fibril orientation and moisture content. Consequently, it is possible to calculate the elastic constants and moisture expansion coefficients of the two solid phases of the mosaic model shown in Fig. 1. The elastic constants of the voids are assumed to be zero.

The phase geometries required for the mosaic model are determined by the volume fractions of bonded material, free fiber segments, and voids, as calculated in Part 2.

Finite element modeling

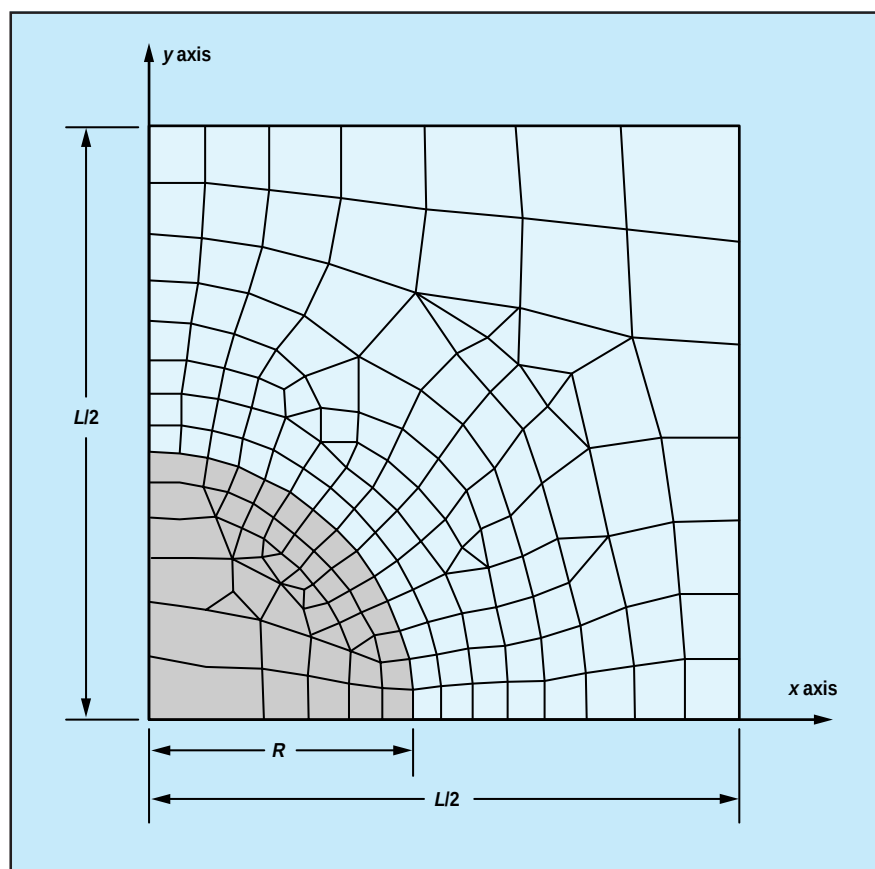
The microstructure of paper has to be specified before the mosaic model can be implemented. However, the fiber segments, fiber crossings, and voids are distributed throughout a paper sheet in a highly complex manner, making a point-by-point description of the structure virtually impossible. To simplify the geometrical modeling of the material, we initially consider paper composed of bonds and fiber segments only.

In a paper of bonds and fibers only, the elastic constants of the bonded material are different from those for the free segments but are within the same order of magnitude (1). There-

fore, the description of the phase geometry should be less critical to prediction accuracy than it would be if the phase properties were highly dissimilar.

Figure 2 shows the simplified geometric description used for the mosaic modeling of paper. The distributed phase is assumed to be of cylindrical shape and to be regularly distributed throughout the volume. As demonstrated in Part 2, the volume fraction of bonds dominates over that of free segments for a sheet of an apparent density above 500 kg/m³. Therefore, it is assumed that bonded material constitutes the continuous phase and that free segments constitute the distributed phase. The simplified mosaic representation of the paper structure is justified by virtue of its computational simplicity alone. Its merits have to be judged on the basis of its ability to predict paper properties.

3. Finite element model of a quarter section of the representative volume element



Since the structure is periodic, it is sufficient to model the representative volume element, depicted in the blowup in Fig. 2. The elastic constants and moisture expansion coefficients of the constituents (fiber segments and fiber crossings) were determined using the series and parallel models.(1)

A two-dimensional, plane stress, finite element model of the representative volume element was constructed to determine the effective in-plane engineering constants. As a result of geometrical and loading symmetries, it is necessary to model only one-fourth of the representative volume element. **Figure 3** illustrates this model.

The four-noded, quadrilateral, isoparametric element STIFX24 was used in the finite element analysis code ANSYS (6). For the purpose of capturing the steep stress and strain gradients at the circular phase

boundary, a fine mesh was constructed near the boundary. The dimensions of the elements at the boundary are $L/40 \times L/40$, where L is the side length of the representative volume element. The total number of element is 172.

The volume fractions of fiber segments and bonded material, V_{segs} and V_{bonds} , in a sheet of paper of a given apparent density are obtained from the Monte Carlo simulation of the paper structure presented in Part 2. The volume fractions of bonded material and fiber segments are different in a sheet of paper and in the representative volume element, because the representative volume element contains no voids. The volume fractions of fiber segments and bonds in the representative volume element, denoted by $V_{\text{RVE,segs}}$ and $V_{\text{RVE,bonds}}$, are:

$$V_{\text{RVE,segs}} = V_{\text{segs}} / (V_{\text{segs}} + V_{\text{bonds}}) \\ = \pi D^2 / 4L^2 \quad (1a)$$

$$V_{\text{RVE,bonds}} = V_{\text{bonds}} / (V_{\text{segs}} + V_{\text{bonds}}) \\ = 1 - V_{\text{RVE,segs}} \quad (1b)$$

where D is the diameter of the cylindrical phase region, as shown in Figs. 2 and 3. The appropriate diameter D is required for finite element analysis of a representative volume element containing certain volume fractions of bonds and segments. D can be obtained by rearranging Eq. 1a:

$$D = (2L / \pi^{1/2}) [V_{\text{segs}} / (V_{\text{segs}} + V_{\text{bonds}})]^{1/2} \quad (2)$$

Calculations of effective engineering constants

Calculation of elastic constants

The in-plane elastic constants of the mosaic model are calculated using the finite element method by imposing appropriate boundary conditions on the representative volume element. To calculate the elastic modulus along the x axis, E_x , we place tensile loading and boundary conditions on the representative volume element.

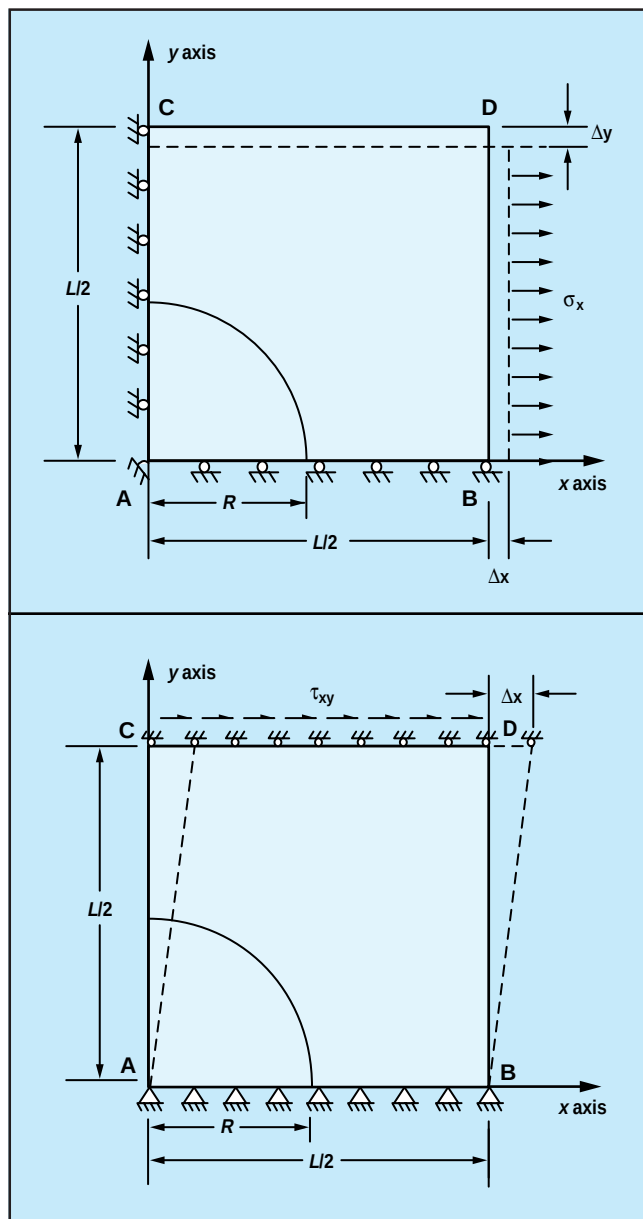
The upper graph in **Figure 4** depicts these conditions. A uniform stress, σ_x , is applied along the boundary indicated in the figure as the "DB" line. This DB boundary is constrained to remain straight after deformation to guarantee compatibility with neighboring representative volume elements. Compatibility with these neighboring volume elements also requires that the boundary denoted "CD" remains straight and that it remains, on average, stress free.

Finite element analysis yields displacements of the boundaries of the representative volume element. The average strain, ϵ_x is

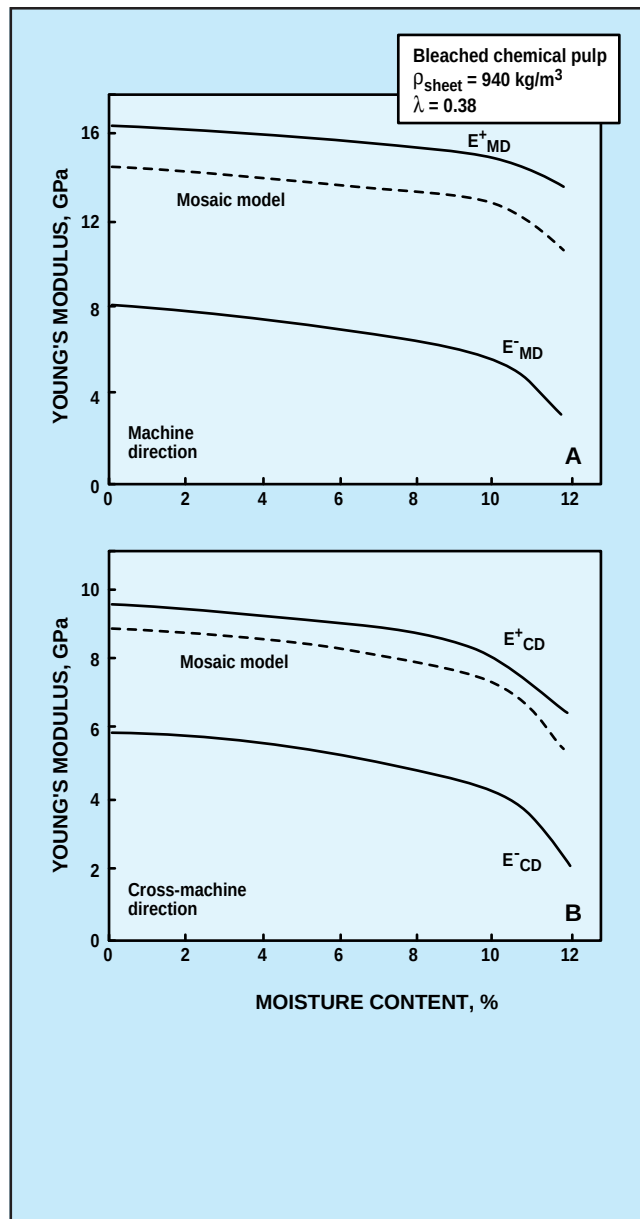
$$\epsilon_x = 2\Delta x / L \quad (3)$$

where Δx is the x -axis displacement of DB boundary and L is the side length of the representative volume

4. Tensile loading and boundary conditions (top) and shear loading and boundary conditions (bottom) applied on the representative volume element



5. Upper and lower bounds and mosaic model predictions of Young's moduli vs. moisture content. (Papers made from bleached chemical pulp. $\rho_{\text{sheet}} = 940 \text{ kg/m}^3$. $\lambda = 0.38$.)



element. Assuming that paper is a macroscopically homogeneous material, the effective elastic modulus, E_x , is

$$E_x = \sigma_x / \epsilon_x \quad (4)$$

Poisson's ratio ν_{xy} can be calculated by

$$\nu_{xy} = \Delta y / \Delta x \quad (5)$$

where Δy is the y -axis displacement of the boundary depicted "CD". The

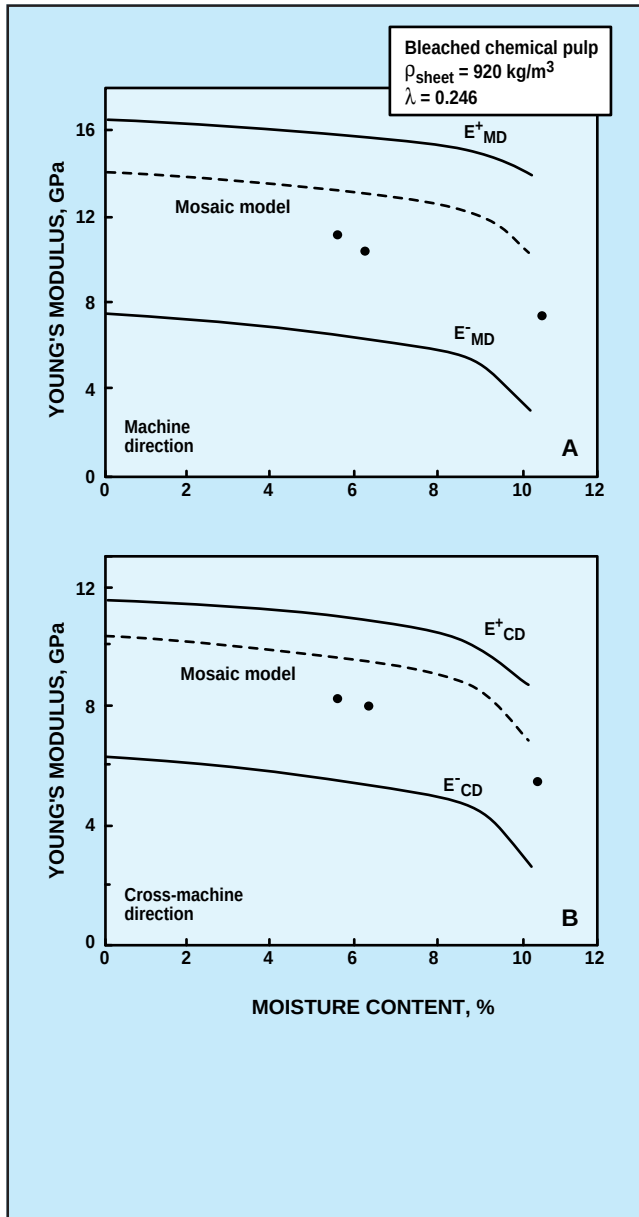
effective Young's modulus, E_y , is similarly calculated by the application of a stress, σ_y .

Boundary conditions appropriate for the calculation of shear modulus are illustrated in the lower graph in Fig. 4. The AB boundary is constrained in both the x and y directions, whereas the CD boundary is constrained to remain straight and horizontal. The AC and BD boundaries should remain straight and parallel during deformation to maintain compatibility with the neighboring representative volume elements after shear deformation.

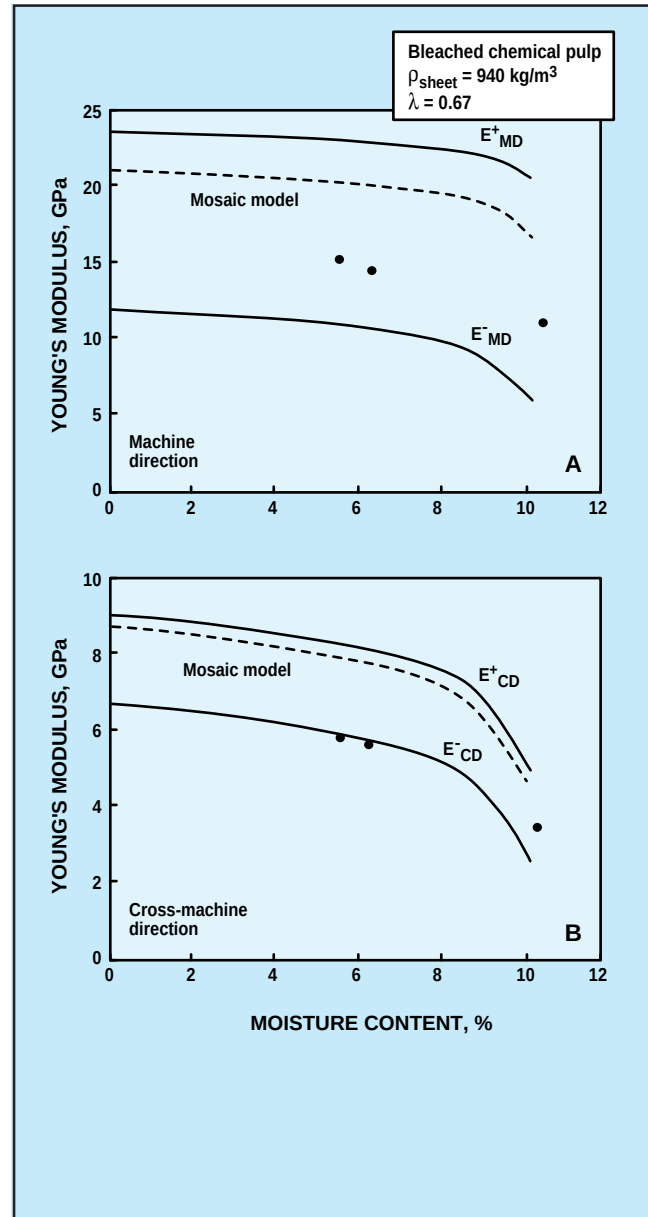
This condition is met by imposing suitable constraints on the nodal displacements. A uniform shear stress, τ_{xy} , was applied on the CD boundary. The average shear strain, γ_{xy} , is

$$\gamma_{xy} = 2\Delta x / L \quad (6)$$

6. Predicted and measured Young's moduli in the machine and cross-machine directions for sheets made from bleached chemical pulp. $\rho_{\text{sheet}} = 920 \text{ kg/m}^3$. $\lambda = 0.246$. Experimental data are from Andersson (10).]



7. Predicted and measured Young's moduli in the machine and cross-machine directions for sheets made from bleached chemical pulp. $\rho_{\text{sheet}} = 940 \text{ kg/m}^3$. $\lambda = 0.67$. Experimental data are from Andersson (10).]



where Δx is the x -axis displacement of the CD boundary. The effective shear modulus, G_{xy} is

$$G_{xy} = \tau_{xy} / \gamma_{xy} \quad (7)$$

The moduli E_x , E_y , and G_{xy} calculated in this way for a void-free paper are corrected for the presence of voids using results obtained for cylindrical through-thickness voids (7). There is no correction of Poisson's ratio pertaining to the

presence of voids (7). The normalized elastic and shear moduli can be related to the volume fraction of voids, V_{voids} , by the following expression (7):

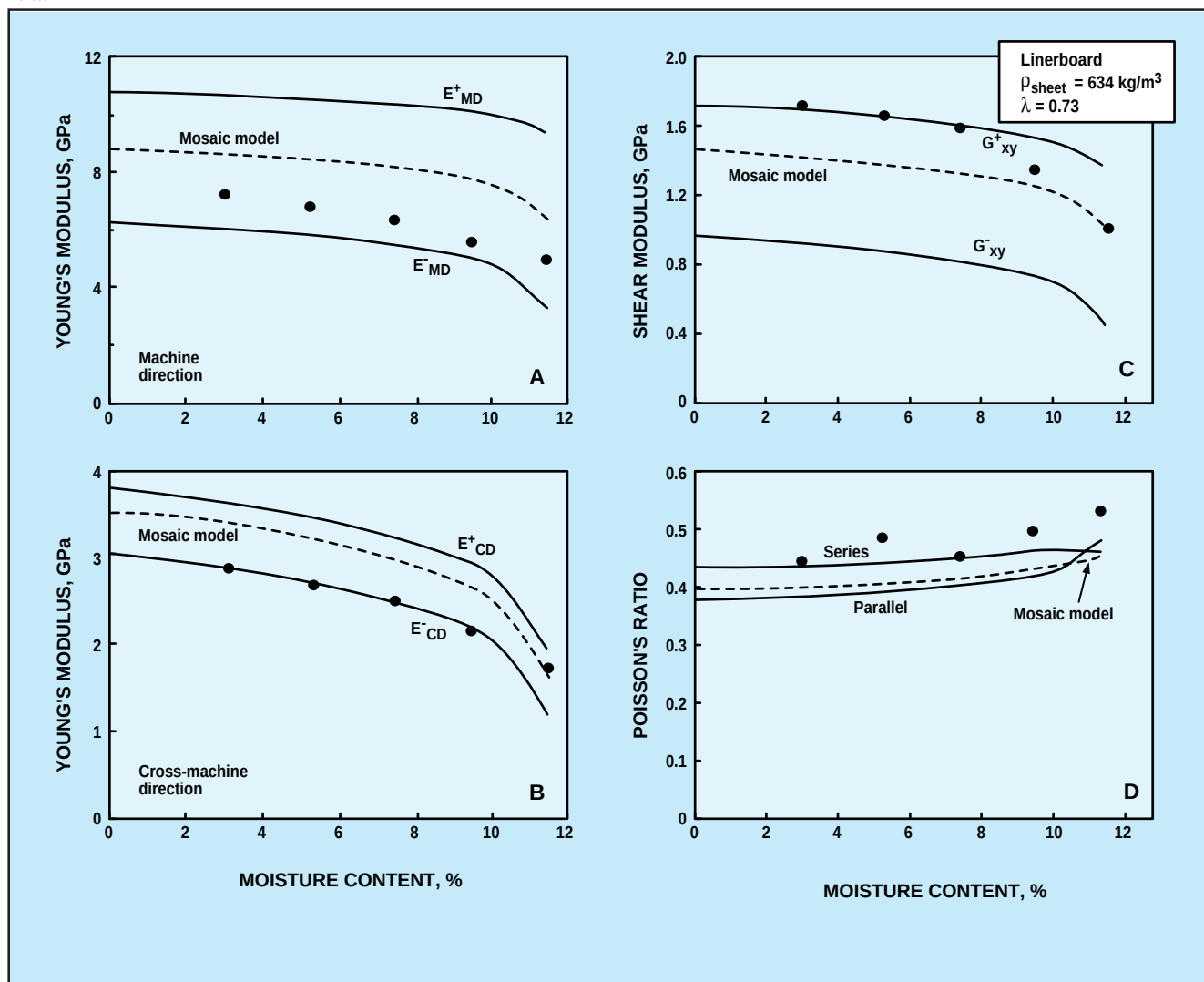
$$y = 1 + \alpha_1 V_{\text{voids}} + \alpha_2 V_{\text{voids}}^2 + \alpha_3 V_{\text{voids}}^3 \quad (8)$$

where $y = E_x/E_{x0}$, E_y/E_{y0} , or G_{xy}/G_{xy0} . E_{x0} , E_{y0} , and G_{xy0} are the elastic and shear moduli of the material without voids (calculated above). E_x , E_y , and

G_{xy} are the corresponding quantities corrected for voids.

The parameters α_1 , α_2 , and α_3 are tabulated elsewhere (7) for different modulus ratios (E_{x0}/E_{y0}) (corresponding to a certain value of the orientation parameter λ defined in Parts 1 and 2). The void volume fraction in a paper sheet, V_{voids} , can be calculated from the apparent density, ρ_{sheet} , of the paper sheet (8):

8. Predicted and measured Young's moduli in the machine and cross-machine directions, shear modulus, and Poisson's ratio of linerboard. $\rho_{\text{sheet}} = 634 \text{ kg/m}^3$. $\lambda = 0.73$. Experimental data are from Baum *et al.* (12).]



$$V_{\text{voids}} = 1 - \rho_{\text{sheet}} / \delta_{\text{cell}} \quad (9)$$

(where ρ_{sheet} is the density of cell-wall material $\rho_{\text{cell}} = 1500 \text{ kg/m}^3$) (9).

Calculation of the moisture expansion coefficients

So far, we have focused on the calculation of elastic constants using the mosaic model. The calculation of the moisture expansion coefficients of paper does not require a mosaic model, inasmuch as Part 1 demonstrated that the upper and lower bounds on moisture expansion coefficients, β_x and β_y , practically coincide.

Furthermore, β_x and β_y of the cell-wall assemblages are unaffected by the voids and pores that are always present in a sheet of paper (7). There is thus no need to correct β_x and β_y for the presence of voids.

Results and discussion

The calculation of the hygro-elastic constants of paper starts with micromechanical calculations of the elastic constants of the cell-wall composed of cellulose, hemicellulose, and lignin, as described in Part 1. The elastic constants of free fiber segments and fiber crossings are re-

spectively calculated from the series and parallel models (1).

After these phase properties of the mosaic model are identified, we turn to the results from the Monte Carlo-simulated sheets of Part 2. These results are employed to identify the appropriate volume fractions of segments and bonds (V_{segs} and V_{bonds}) corresponding to any given apparent sheet density.

Based on this information, it is possible to calculate the appropriate dimensions of the representative volume element (Eq. 2). Finally, the effective engineering constants of paper are obtained from finite element analysis of the representative

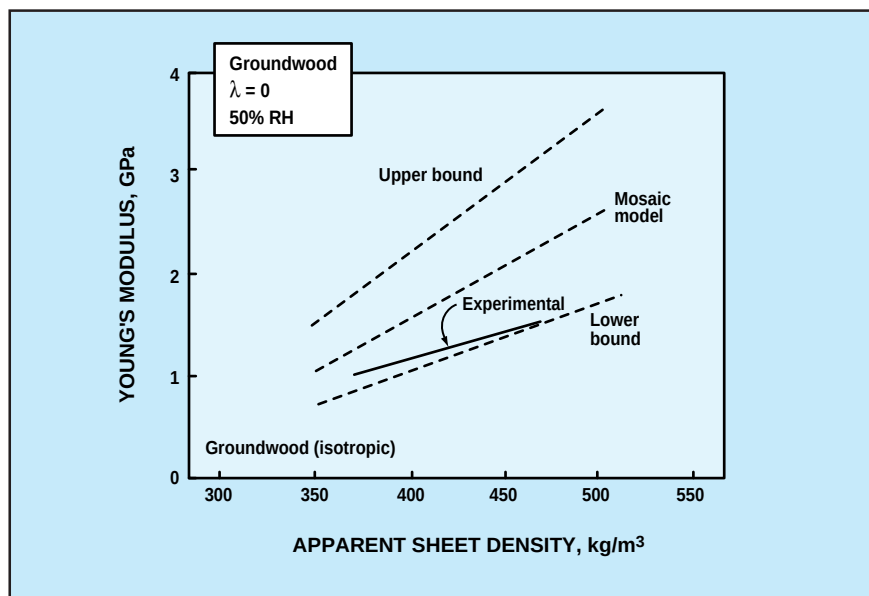
I. Measured (9) and calculated moisture expansion coefficients for papers made from bleached chemical pulp

	β_{MD}	β_{CD}
Isotropic sheet		
Experimental	0.11	0.11
Predicted	0.19	0.19
Oriented sheet		
Experimental	0.06	0.22
Predicted	0.05	0.33
$\rho_{\text{sheet}} = 800 \text{ kg/m}^3$.		

II. Measured (10) and calculated moisture expansion coefficients for paper made from bleached chemical pulp

	β_{MD}	β_{CD}
Low-anisotropy paper ($E_{MD}/E_{CD} = 1.4$, $\rho_{\text{sheet}} = 920 \text{ kg/m}^3$)		
Experimental	0.14 ± 0.09	0.22 ± 0.11
Predicted	0.13	0.20
Highly oriented paper ($E_{MD}/E_{CD} = 2.6$, $\rho_{\text{sheet}} = 940 \text{ kg/m}^3$)		
Experimental	0.11 ± 0.06	0.38 ± 0.16
Predicted	0.06	0.26

9. Predicted and measured Young's moduli vs. apparent sheet density for isotropic ground-wood sheets. [50% RH. $\lambda = 0$. Experimental data are from Fellers *et al.* (14).]



volume element, as discussed in the previous section.

In examining how well the model predicts the properties of paper, we will consider papers made from bleached chemical pulp, linerboards, and papers made from groundwood pulp fibers.

Bleached chemical pulp

Figure 5 shows mosaic model results plotted against moisture content. The upper and lower bounds are also shown, indicated by plus and minus superscripts on E_{MD} and E_{CD} . These papers were made from bleached chemical pulp, at 55% cellulose and 45% hemicellulose (by vol-

ume) (9), with an apparent density $\rho_{\text{sheet}} = 940 \text{ kg/m}^3$ and orientation parameter $\lambda = 0.38$. Both E_{MD} and E_{CD} decrease with increasing moisture content as a result of the softening of hemicellulose and lignin.

As expected, the mosaic model predictions fall between the bounds, but they are closer to the upper bound than to the lower bound. This outcome can be accounted for by the differences in volume fractions. In such a high-density paper, the bonded material dominates over free segments.

Andersson performed measurements of Young's moduli in the machine and cross-machine directions

for papers made from bleached chemical pulp ($\rho_{\text{sheet}} = 920$ and 940 kg/m^3) over a range of relative humidities (10). The chemical composition of the pulp fibers was 55% cellulose and 45% hemicellulose (by volume) (9). The average fiber length, width, and thickness were 1.96 mm, 19.3 μm , and 5 μm , respectively (10).

The orientation parameter λ in the Cauchy distribution function (1, 2) was not measured. However, we were able to back-calculate the value by comparing and adjusting λ until the elastic modulus ratio E_{MD}/E_{CD} calculated from the mosaic model agreed with the experimental value. For sheets of densities $\rho_{\text{sheet}} = 920 \text{ kg/m}^3$ and 940 kg/m^3 and modulus ratios $E_{MD}/E_{CD} = 1.4$ and 2.6 at 50% RH (10), this procedure yields $\lambda = 0.246$ and 0.670, respectively.

Figure 6 shows a plot of measured and calculated Young's moduli for the low-anisotropy paper ($\lambda = 0.246$) vs. moisture content. The predictions of the moduli E_{MD} and E_{CD} from the mosaic model are somewhat high although they are in reasonable agreement with the experimental modul. The papers with which Andersson worked were dried under constraints but were subsequently subjected to cyclic humidity changes between 30% and 90% RH to relieve dried-in stresses. This process is known to reduce the moduli by the introduction of "microcompressions" at the bond sites (11). Our model predictions are

made under the assumption that the cell wall contains no micro-compressions, which would explain the overprediction of the moduli.

Figure 7 shows E_{MD} and E_{CD} plotted against moisture content for the more highly oriented paper ($\lambda = 0.67$, $\rho_{sheet} = 940 \text{ kg/m}^3$). The mosaic model again overpredicts the moduli E_{MD} and E_{CD} . The discrepancies between experimental and predicted values are attributed to the factors already discussed.

The moisture expansion coefficients β_{MD} and β_{CD} can be calculated from either the series model or the parallel model (1). Let us consider first the experimental data for bleached chemical pulp published by Salmen *et al.* (9). They examined isotropic and oriented sheets of a density of 800 kg/m^3 . To compare our model and their data, we adjusted the orientation parameter λ until the calculated and measured modulus ratio E_{MD}/E_{CD} for the sheets coincided ($E_{MD}/E_{CD} = 1.0$ for $\lambda = 0$ and 3.0 for $\lambda = 0.812$).

Table I summarizes measured and calculated moisture expansions coefficients β_{MD} and β_{CD} . As the table shows, the model overpredicts β for the isotropic sheet ($\lambda = 0$), while the predictions are more reasonable for the oriented sheet ($\lambda = 0.812$). The reasons for the discrepancies are not known, but the differences may possibly be attributed to the micromechanical modeling and to the assumption of isotropic swelling and volume additivity used for the hemicellulose (1). Furthermore, the amorphous fraction of cellulose is considered as being a part of the hemicellulose, which may not be appropriate when predicting moisture expansion.

Table II compares moisture expansion coefficients measured on sheets made from bleached chemical pulp (10) and calculations from the model. The table includes the paper of low anisotropy of Fig. 6 and the highly oriented paper of Fig. 7. The predicted hygroexpansion coefficients β_{MD} and β_{CD} are in reason-

able agreement with experimental data, but the large scatter prohibits detailed comparisons.

Linerboard

Baum *et al.* measured the in-plane elastic constants of linerboard of apparent density of 600 kg/m^3 between 20% and 80% RH (12). The chemical composition of the fibers, fiber dimensions, and fiber orientation data were not provided in their study. For the prediction of mechanical properties, the fibers in linerboard were assumed to contain 40% cellulose, 46% hemicellulose, and 14% lignin (by volume) (13). The fiber length, width, and thickness were assumed to be 2 mm, $25 \mu\text{m}$, and $5 \mu\text{m}$, respectively (13). The parameter λ is 0.73, according to the back-calculation procedure.

Predicted curves and experimental data on the Young's moduli, shear modulus, and Poisson's ratio for linerboard are shown in **Fig. 8**. Both E_{MD} and E_{CD} calculated from the mosaic model somewhat exceed the experimental data. Figure 8C shows the results for the shear modulus G_{xy} . At low moisture contents, the experimental data on G_{xy} almost coincide with the upper bound. At high moisture contents, the mosaic model predictions of the shear modulus are very close to the measured experimental data. Figure 8D shows that the upper and lower bounds on Poisson's ratio ν_{xy} are quite close. The mosaic model slightly underestimates the Poisson's ratio ν_{xy} .

Sheets from groundwood pulp

The elastic modulus of isotropic paper sheets made from groundwood pulp was calculated for a range of apparent densities at 50% RH. Groundwood pulp was assumed to contain 36% cellulose, 40% hemicellulose, and 26% lignin (by volume) (13). The fiber length, width, and thickness were assumed to be 1 mm, $22 \mu\text{m}$, and $5 \mu\text{m}$, respectively.

Figure 9 shows a plot of calculated and measured Young's moduli

vs. apparent density, ρ_{sheet} . The solid line is based on the experimental results presented by Fellers *et al.* (14), and the dotted lines are predictions we made using the mosaic model. The measured modulus is close to the lower bound.

In groundwood pulp, only about 20% of the solids content consists of long fibers. Groundwood pulp contains about 30% fines and 50% fibers in the middle fraction (15), which would tend to reduce the modulus of the sheet. The model presented here does not take into account the way in which fines may contribute to load transfer, nor would it account for the substantial variations in fiber dimensions in such a sheet.

The model would thus need to be modified for papers made from mechanical pulps, especially those made from groundwood pulp, which contains only a small fraction of long fibers. Moreover, the apparent density of groundwood pulp sheets is quite low, and continuum modeling of such sheets may be questionable.

Conclusions

This mosaic model can be used to calculate the elastic properties of paper, which is considered to be composed of free fiber segments, fiber crossings, and voids. The most important parameters for the use of the mosaic model are apparent sheet density, moisture content, and fiber orientation. Also required are fiber dimensions, but they are of minor importance according to the analysis presented in Part 2. The chemical composition of the cell wall (volume fractions of cellulose, hemicellulose, and lignin) is also important, particularly because the elastic constants are sensitive to moisture.

The finite element method is used to determine effective hygro-elastic properties of paper, based on analysis of the representative volume element. After correcting the elastic stiffnesses for voids in the paper, the predictions of elastic constants can be compared with experimental

data. The prediction of the moisture expansion coefficients does not require the mosaic model because the lower and upper bounds practically coincide.

The predictions of elastic moduli in the machine and cross-machine directions for papers made from bleached chemical pulp and for linerboards were in reasonable agreement with experimental data collected over a wide range of moisture contents. The mosaic model predictions of the elastic moduli were generally higher than the values observed in the experiments. This result may be attributable to microcompressions in fiber crossings or curved fiber segments, which are factors not covered by the model.

The predicted shear modulus of linerboard was below the experimental data at low humidities and was in very good agreement with experimental values at high humidities. The experimental shear modulus was thus more sensitive to the softening influence of moisture than the predicted shear modulus.

The predictions of Poisson's ratio were in good agreement with experimental data.

The predicted Young's modulus for isotropic groundwood sheets was larger than that observed experimentally. This outcome may arise from the presence of materials other than long fibers and from the low apparent density that such papers have.

Finally, the composite model provides reasonable predictions of moisture expansion coefficients, but substantial experimental scatter prevents more detailed comparisons. More accurate experimental data are needed to further judge the merits of the modeling. The regular arrangement of free fiber segments and bonded material in the mosaic model is a very primary simplification. A more realistic geometrical representation of the microstructure of paper would probably give more accurate predictions of paper mechanical properties. ■

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