Determination of Paper Sheet Fiber Orientation Distributions by a Laser Optical Diffraction Method

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Synopsis

The combination of the laser and optical diffraction analysis provides a powerful tool for the determination of fiber orientation distribution in any paper sheet of basis weight low enough to be translucent to the laser beam. The coherent light beam from the laser is diffracted by the sample. It undergoes a Fourier transform from which the orientation distributions can be calculated. Measurements were made on 14 different samples of various degrees of orientation, including an essentially random handsheet as a control. The cross-direction/machine direction elastic modulus ratios of the samples were found to correlate well with the distribution functions fitted to the orientation data. The correlation curve was S-shaped, with a drastic drop in the modulus ratio in the medium range of fiber orientation.

INTRODUCTION

Most commercially produced fiber network systems are anisotropic in terms of their structure and material properties. Machine-made paper and nonwoven fabrics are particular examples. The structure-property relationships reflect this anisotropy in many ways. The solutions of analytical approaches to predicting mechanical properties depend on accurate and reasonably rapid techniques for measuring fiber orientations.

A commonly employed technique involves making sheets with a small fraction of dyed fibers in the furnish, then determining the orientation of the dyed fibers microscopically.¹⁻⁵ Since a large number of fibers must be examined to obtain meaningful results, these methods become tedious and time consuming. Kinks or curves in the fibers complicate the analysis; they cannot be treated as individual straight lines. Also there may be differences in orientation between the felt and wire sides; observations limited to the two surfaces may not provide a valid analysis of the overall orientation distribution.

Other methods have been used or proposed by various researchers. X-ray diffraction⁶⁻⁸ yields quantitative data but requires complex instrumentation and it is necessary to measure the fibril angle of the sample material independently. Small-angle light scattering⁹ is a powerful method for strongly broken-up or regenerated cellulosic material but is less effective for papers in which the fibers are relatively intact. Zero-span tensile ratios¹⁰ are relatively

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simple and rapid, but the results are in terms of strength anisotropy, which is known to be affected by factors other than structural anisotropy. Line intersection methods, which are strictly statistical in nature, have also been applied.¹¹

For a more thorough description and comparison of the above methods, the reader is referred to Mark.¹² This reference also discusses theoretical orientation distributions, and methods of fitting data to them.

Optical diffraction analysis has been successfully applied to extract useful information concerning orientation trends in contour maps and related charts, particularly in the earth sciences.¹³⁻¹⁷ Distributions of trends in topography, pressure, temperature, population density, geomagnetic intensity, etc., are determined from such maps for use in spatial analysis. Applications of this method to other types of two-dimensional displays, chiefly photographs, have been demonstrated.¹⁸⁻²⁰ The goal of this study was to employ optical diffraction analysis as a means of fast, accurate determination of fiber orientation distribution in paper and nonwoven sheets.

METHOD OF ANALYSIS

Fourier Transform

A parallel beam of light is called coherent if all of its rays, when crossing any plane perpendicular to the beam, have the same phase. A coherent beam remains coherent after passing through an object of uniform thickness, and if such a beam is focused to a point by a lens, the rays will still have the same phase when they arrive at the focal point. With coherent optics, the best source of illumination is the continuous wave laser.

Let a translucent sample (e.g., a thin sheet of paper) A in Figure 1 be illuminated by a coherent beam of light of wavelength λ parallel to the optic axis. Thus, each ray of the beam incident on the plane of A has the same amplitude E_o and phase ψ_o . Let the ray passing through A at the point (x, y)be attenuated in amplitude by the factor f(x, y) and retarded in phase by





 $\psi(\mathbf{x}, \mathbf{y})$ so that the amplitude and phase of the ray as it emerges from A are $f(\mathbf{x}, \mathbf{y})E_{o}$ and $\psi(\mathbf{x}, \mathbf{y}) + \psi_{o}$, respectively.

On passing through A, the light is diffracted. Let us consider that part of the light which is diffracted at a given angle, having direction cosines A1 and A2 with respect to the x and y axes. This parallel family of light rays is focused by the lens to a single point in the focal plane, whose coordinates ω_x and ω_y are proportional to A1 and A2, the factor of proportionality being L/A3, where $A1^2 + A2^2 + A3^2 = 1$ and L is the focal length of the lens. Let P be the plane through the center of A whose normal has direction cosines A1 and A2. Then, the phase of the ray through (x, y) when it crosses P is $\psi_a + \psi(x, y) + 2\pi(A1x + A2y)/\lambda$.

The phase of the ray through (x, y) when it arrives at (ω_x, ω_y) differs by a constant from its phase when it crossed *P*. If diffraction at large angles is assumed to be negligible, we may suppose that A3 \approx 1 is independent of (ω_x, ω_y) . The sum of the rays at (ω_x, ω_y) is proportional to the quantity

$$\int \int f(\mathbf{x},\mathbf{y}) e^{i\psi(\mathbf{x},\mathbf{y})} e^{2\pi i(\omega_{\mathbf{x}}\mathbf{x}+\omega_{\mathbf{y}}\mathbf{y})/L\lambda} d\mathbf{x} d\mathbf{y}$$

which becomes the Fourier transform of the complex function

$$f(\mathbf{x},\mathbf{y})e^{i\psi(\mathbf{x},\mathbf{y})}$$

scaled by the factor $1/L\lambda$.^{21,22}

When parts of a beam of coherent light are blocked, the beam is diffracted, and the amplitude and phase of the diffracted light depend on the pattern of points at which the beam is blocked. Photographic film is a square-law detector, recording light intensity but not the phase differences which are present. The light intensity in the Fourier transform focal plane is the square of the amplitude of this Fourier transform (i.e., the power spectrum of A).

Optical Diffraction Analysis

In this analysis a fiber network is viewed as a spatial system of curved and/or linear elements in two dimensions. The significant properties of the elements are their orientation and spatial frequency.

The optical diffraction method is based on principles first set forth in 1873 by Abbé (see Ref. 23). The appearance of the laser in 1960 rendered the method practical. It provides a means for analyzing information presented two-dimensionally, as in the form of photographs, drawings, and maps.

Rudström and Sjölin²⁴ were the first to describe the application of optical diffraction analysis to the determination of fiber orientation distribution. Sadowski²⁵ later presented a slightly different theoretical development. Of these two, the former present no results at all, while Sadowski's results are general and qualitative.

The basic technique involves spectral analysis of the sample's spatial (geometric) properties. The sample is either a translucent material or a transparent replica of some convenient scale, typically 35 mm film, that functions as a diffraction grating with initially unknown spatial properties. On

the other hand, the spectral properties of the source of illumination are precisely known, that is, a laser provides coherent monochromatic light. In mathematical terms, the resulting diffraction pattern is the two-dimensional Fourier transform of the complex amplitude of the transmitted light. In simpler terms, the transform is a plot of the distribution of orientations and spatial frequencies of the elements in the specimen.

By use of diffraction patterns or transforms, various sheet samples can be compared quantitatively in terms of fiber element spacing, orientation distributions, and symmetries, regardless of scale.

EXPERIMENTAL

Laser Equipment

The laser system used in this study is a C-120 Laser Scan, shown in Figure 2, in which light from a ruby laser source is emitted at 632.8 nm. Output power was 6 mw. With additional optics, the original image can be reconstructed from the light rays that form the diffraction pattern.

Materials

Three types of nonwoven materials were examined: natural fiber nonwovens, nonwovens made with cellulose derivatives, and synthetic fiber nonwovens. A handsheet of wood pulp was also examined to serve as a reference material with random fiber orientation. Because the test specimen material had to be quite translucent, only light- and medium-weight materials were used. A total of 14 samples were examined; their fiber components, where known, were listed in Table I.

Mechanical Testing

After the samples were equilibrated at TAPPI standard temperature and humidity conditions (73°F, 50% relative humidity), specimens were prepared in the machine and cross-machine directions, and tested in tension to obtain modulus of elasticity E. A linear variable differential transformer (LVDT) was used to measure the displacements independently from the crosshead movement of the testing machine.

Photography of Fourier Transform

A 1.5×3 inch specimen was cut from each sample, with the long axis oriented in the machine direction. The specimen was placed in the film gate assembly of the laser bench. A circular light trap frame assembly was placed between the collimator lens and the specimen to block out the light and to yield an incident coherent light beam one inch in diameter. A 4×5 inch Polaroid film, type 55 P/N was positioned in the focal plane of the Fourier transform. The film recorded the light intensity of the Fourier transform showing the orientation and spatial pattern of the fiber elements within a one-inch circular area.



Fig. 2. The LaserScan C-120 Apparatus. (A) Laser light source, (B) beam-expanding telescope, (C) collimating lens, (D) sample holder & gates, (E-E) focusing lenses, (F) TV camera in transform plane, (G) TV monitor, (H) filmholder, (J) optical bench. Camera F and monitor G are used for setup and focusing. The camera is then swung out of the light path and the filmholder moved up to the transform plane. Reprinted from Ref. 12, p. 300, by courtesy of Marcel Dekker, Inc.

Sample no.	Material ^a	Basis wt., g/m	Density, g/cm	$E_C/E_M^{\rm b}$	Orientation parameter D2
Natural f	ibers:				
1.	Slash pine handsheet	20.0	0.328	1	0
2.	Lens paper (wood pulp)	8.4	0.220	0.29	0.055
3.	Nonwoven:100% cotton	76.4	0.430	0.53	0.033
Cellulose	derivative nonwovens:				
4.		24.8	0.271	0.08	0.055
5.	(ca. 50% wood pulp)	22.0	0.247	0.70	0.032
6.		59.3	0.243	0.85	0.037
7.		25.1	0.282	0.08	0.067
8.		39.3	0.245	0.17	0.050
9.	(contains small amount	26.8	0.245	0.05	0.047
Sunthatia	of wood pulp)				
10	100% Polyester	56.8	0.369	0.34	0.052
10.	100% Polyester	J0.0	0.302	0.04	0.002
11.	50% Polyester	44.0	0.227	0.02	0.021
14.	50% nylon	47.0	0.215	0.02	0.070
13.	100% Polyester	68.2	0.323	0.18	0.039
14.	30% Polyester, 70% nylon	55.5	0.263	0.04	0.055

TABLE I Properties of Sample Materials

^aSample material was supplied by Stacy Fabrics Corporation, Johnson & Johnson, and Staple Fabrics Corporation. We gratefully acknowledge their assistance.

 ${}^{b}E_{C}$ = Modulus of elasticity in cross-machine direction.

 E_M = Modulus of elasticity in machine direction.

Quantification of the Fiber Orientation Distribution

A light densitometer was used to scan the light intensity of the Fourier transform film negatives at ten-degree intervals (from the minor axis $[0^{\circ}]$ to the major axis $[90^{\circ}]$ of the diffraction pattern). The ten continuous x-y plots were analyzed with the aid of a point mode digitizer. Since the transform is a plot of the distribution of orientations and spatial frequencies of the fiber elements in the specimen material, the ratio of the area of an x-y plot to the total area measured is approximately equivalent to the fraction of fiber orientation lying at that angle. The fiber orientation distribution is determined in this manner.

RESULTS AND DISCUSSION

Results of Mechanical Tests

The ratio of modulus of elasticity in the cross-machine direction to that in the machine direction was calculated for each sample; these figures are given in Table I, along with basis weights and densities.

Orientation Distribution of Fiber Elements

Figures 3, 4, and 5 illustrate results obtained with fiber network samples and their corresponding Fourier transforms. A network with random fiber



(a)



(b)

Fig. 3. (a) A randomly oriented fiber network (Sample no. 1) (90 \times). (b) Fourier transform for this network.



(a)



(b)

Fig. 4. (a) A slightly oriented fiber network (Sample no. 5) $(90 \times)$. (b) Fourier transform for this network. The arrow indicates the plane of maximum scattered light perpendicular to the primary direction of fiber orientation.

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(a)



(b)

Fig. 5. (a) A highly oriented fiber network (Sample no. 12) $(90 \times)$. (b) Fourier transform for this network. The arrow indicates the plane of maximum scattered light perpendicular to the primary direction of fiber orientation. Reprinted from Ref. 12, p. 300, by courtesy of Marcel Dekker, Inc.

orientation, as in Figure 3, has a circular light scattering pattern. On the other hand, a network with oriented (anisotropic) fiber alignments has an elliptical light scattering pattern, with the minor axis of the Fourier transform corresponding to the direction of maximum orientation of the fibers in the sheet. Figures 4 and 5 show slightly and highly oriented networks, respectively. Miles²⁶ developed a function that has been used to describe nonrandomly oriented fiber networks.²⁷⁻²⁹ The function has the form

$$F(\theta) = 1/\pi - b1\cos(2\theta) \tag{1}$$

where $F(\theta)$ is the probability that a fiber lies at an angle θ to the crossmachine direction. The factor b1 is related directly to the fiber orientation distribution.

On integration, Eq. (1) meets the requirement

$$\int_0^{\pi} F(\theta) \, d\theta = 1$$

For a randomly oriented network, b1 = 0 and $F(\theta) = 1/\pi$.

Equation (1) is modified in this study to fit the experimental data of fiber orientations by the expression

$$F(\theta) = D1(1/\pi - D2\cos(2\theta))$$
⁽²⁾

where D1 is a parameter that depends on the number of (equal) sectors selected for scanning over a specified circular arc and D2 is an orientation distribution parameter. D1 and D2 are determined by fitting Eq. (2) to the experimental data by a least-squares method. D2 values are given in Table I; the higher the value, the greater is the anisotropy of the fiber network. D1 is equal to about 0.314 for all cases in this study.

Figure 6 represents the fiber orientations for three samples of widely different degrees of orientation. Symbols indicate the experimental data points and the lines are obtained by applying Eq. (2). Since the light scatter-



Fig. 6. Fiber orientation distributions for samples of different degrees of orientation: Random (Sample no. 1), slightly oriented (Sample no. 5), and highly oriented (Sample no. 12). (×) Sample #1 $D_2 = 0$; (•) Sample #5 $D_2 = 0.032$; (\bigcirc) Sample #12 $D_2 = 0.076$.



Fig. 7. The relationship between CD/MD modulus of elasticity ratio E_C/E_M and the orientation factor D2.

ing of the Fourier transform is symmetric, only the first and third quadrants, 0 to 90 and 180 to 270 degrees, were analyzed. Note the excellent agreement between the experimental data and values calculated from Eq. (2). Accordingly, this expression finds ready use in the development of the elasticity theory of nonrandomly oriented fiber networks.

Sheet mechanical properties are related to fiber orientation distributions. A randomly oriented network of the handsheet type has roughly isotropic properties (i.e., the ratio of cross-machine to machine direction modulus of elasticity $E_C/E_M \approx 1$). A machine-made fiber system is usually stronger and stiffer in the machine direction. Figure 7 shows the relationship between modulus ratio E_C/E_M and the orientation factor D2, along with a line of best fit to the data. The shape of this line somewhat resembles that of one fit to a plot of zero-span E_C/E_M versus x-ray orientation parameter data by Koran et al.³⁰

Since the laser beam must pass through the sample, the basis weight of the sample material is limited by the output power of the laser. In this study the relatively low power (6 mw) restricted the application to materials of relatively low basis weight. A more powerful laser would extend the usefulness of the method to heavier materials.

Several techniques, such as cold-drum sheet splitting³¹ and sheet splitting by peeling layers away with pressure-sensitive tape, were tried in order to obtain acceptably thin layers from thicker samples (sheets too heavy to transmit the beam). These techniques do not produce sample layers of adequate flatness and uniformity to yield a reliable Fourier transform.

It has been suggested that opaque paper samples can be treated with polymers to improve their light-transmitting qualities without disturbing the network structures.³² This approach might warrant further investigation.

It might be possible to combine the laser diffraction system, as described here, with image analysis equipment. Note the television camera in Figure 2; this could be used to feed diffraction patterns directly into appropriate processing equipment.

A possible practical application of this method might be the on-line onmachine monitoring of orientation distribution. A continuous typically elliptical pattern would be output; any change would indicate a difference in the formation.

SUMMARY AND CONCLUSIONS

An optical diffraction analysis technique for measuring orientation distributions of fiber networks has been developed. A laser supplies the coherent light beam on a translucent fiber network, which functions as a diffraction grating. With suitable optics, a Fourier transform of the diffraction pattern was produced. A light densitometer was used to scan the light intensity of the film negatives of the Fourier transform, and fiber orientation distributions were determined.

Orientation measurements were made on 14 light- or medium-weight fiber systems, including three types of nonwovens. A handsheet of wood pulp was included as a reference pattern of a random (isotropic) fiber orientation distribution.

A distribution function with two parameters, used to describe the fiber orientation distributions, gave excellent agreement with the experimental data. A relationship between the orthotropic mechanical properties of the samples and their fiber orientation distributions was demonstrated.

The optical diffraction analysis technique provides a powerful tool for fiber network analysis.

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