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D.H. HORSTMANN, J.D. LINDSAY, AND R.A. STRATTON

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Diane H. Horstmann
Jeffrey D. Lindsay
Robert A. Stratton
The Institute of Paper Science and Technology
Atlanta, Georgia 30318

ABSTRACT

The results from a new experimental method to characterize edge penetration in photographic papers were found to provide information on in-plane permeability. The new method provides a simple way of examining the anisotropic in-plane permeability of paper. The technique employs vacuum-driven lateral injection of fluid in an initially dry, laminated paper disk. Simple image analysis combined with approximate solutions to the flow problem allow permeability results to be obtained from photographs of the moving fluid boundary. Although the in-plane permeability changes with time as the fibers swell, the degree of in-plane anisotropy (defined as the MD-CD permeability ratio) does not appear to change significantly with swelling, consistent with the findings of another recent study.

INTRODUCTION

While flow normal to the plane of paper is of great importance in many papermaking processes, the in-plane flow of liquid through paper is receiving increasing attention. Significant in-plane flows of water during wet pressing can occur[1-3], and lateral penetration of coating colors can be important in blade coating processes[4]. Of particular importance to the photographic industry is edge penetration, which is the lateral infiltration of fluid through the exposed edges of a paper having both surfaces made impermeable to fluid. An otherwise excellent photo can be ruined by significant edgewise penetration of developing solutions, which causes discoloring around the edges of the photograph. The process of edgewise penetration of a fluid is also of concern when defects of "pinholes" may be present in packaging materials for liquids such as milk cartons.

The flow of fluid through paper is largely determined by the permeability of the sheet. The structure of paper ensures that permeability will vary with direction, or in other words, the permeability of paper is anisotropic. Where in-plane flow is concerned, the permeability in the machine direction of the sheet is likely to be different from that in the cross-direction. However, there has been a severe lack of published experimental data on the anisotropic permeability of paper. This paper reports a new, simple technique for measuring in-plane anisotropy of an initially dry disk of paper, supplementing the recently published theoretical and experimental work on anisotropic permeability in paper[5].

Previous Work

Measurements in paper

In the past, in-plane measurements of permeability in paper appear to have been entirely absent from the literature. This lack of data is surprising, since similar measurements have been made in felts, textiles, rocks, and other porous materials. Experimental difficulties in constraining the flow to the plane of the paper, without permitting channeling along the surface of the sheet, seems to have hampered some previous efforts (see discussion by Lindsay[6]).

Recently Lindsay[6] published data on all three components of the permeability tensor for paper. The experimental approach used a disc of paper sealed between two carefully aligned metal platens by the application of uniform hydraulic pressure. Dyed fluid was injected into a small hole in the center of the disc and forced to flow radially outward in the plane of the paper. Injection rates were sufficient to ensure that capillary flows were negligible, thus permitting Darcian permeability to be measured. Numerical and analytical techniques were also developed to use information about the flow rate of the fluid or the size and shape of the dye boundary in obtaining in-plane permeabilities. The ratio of machine-direction to cross-direction permeability was obtained in several papers, with values generally in the range of 1-2, and frequently in the range of 1.1-1.3.

Back has examined in-plane capillary flows in paper[7]. By observing the way in which a drop of fluid spreads out in a sheet of paper, information about anisotropy in the sheet can be obtained. He found that the capillary penetration velocity for most papers is 5-15% larger in the machine direction than in the cross-direction. However, capillary flow is affected by both the intrinsic Darcian permeability of the sheet as well as the (directional) pore size distribution. No quantitative conclusions about permeability can be safely deduced from such experiments unless detailed information about directional pore-size distributions is available.

Recently, the results from a new experimental method to characterize edge penetration in photographic papers[8] were found to provide information on anisotropic in-plane permeability. The information about in-plane anisotropy supplements the results obtained with different methods by Lindsay. The new method, described below, provides a simple and inexpensive way of measuring the two in-plane permeability components of paper.

EXPERIMENTAL

The permeability data reported in this study were obtained as part of a study by Horstmann[8] on edge penetration in paper. The basic approach involved evacuating the air in a dry disk of paper which has been laminated with impermeable plastic, and then allowing liquid at atmospheric pressure to be driven through the edges of the disk toward the center. The penetrating liquid creates a moving boundary between the wet and dry regions of the paper. The shape and size of this boundary can be used to obtain information about the anisotropic in-plane permeability of the sheet.

Fourdrinier neutral sized bleached kraft paper was produced for this study. All papers tested had nominal basis weight of 195 g/m^2. A barrier coating of clear, high density polyethylene was laminated onto both faces of the paper samples at a temperature of 150-155°C, making the surfaces impermeable in order to ensure that flow occurred only in the plane of the paper. Disks of the laminated paper, 7.6 cm in diameter, are punched using a Thwing Albert disk cutter. The samples are then conditioned for a minimum of 48 hours at 50% RH and 22°C.

The penetrating fluid used in these experiments was an aqueous solution used commercially with the base paper described above. The liquid contained 80-85% water, by weight, and had a pH of 11.4. The viscosity at room temperature was 1.90 centipoise. Fresh solution was needed for each trial because it oxidized readily upon exposure to light and air.

Figure 1 shows the test chamber, which consists of a 25-cm wide desiccator jar with a flat Lucite lid. There are two openings in the lid: one for pulling the vacuum and the other for injecting the penetrating liquid. For each run, the laminated disk of paper is attached to the bottom of the test chamber with a piece of tape to prevent floating once the sample is immersed in liquid. Before liquid is added, the air in the sample is removed by applying vacuum pressure (-.96 atm gauge) through one of two ports on the chamber lid. A second port in the lid is used to inject the penetrating fluid. Once the air has been removed from the chamber and the sheet, a valve is opened allowing liquid to enter the chamber and immerse the sample. Atmospheric pressure is quickly achieved as air flows in through the fluid injection valve, and the vacuum line is closed. A driving force of approximately 1 atmosphere thus propels fluid into the evacuated paper.

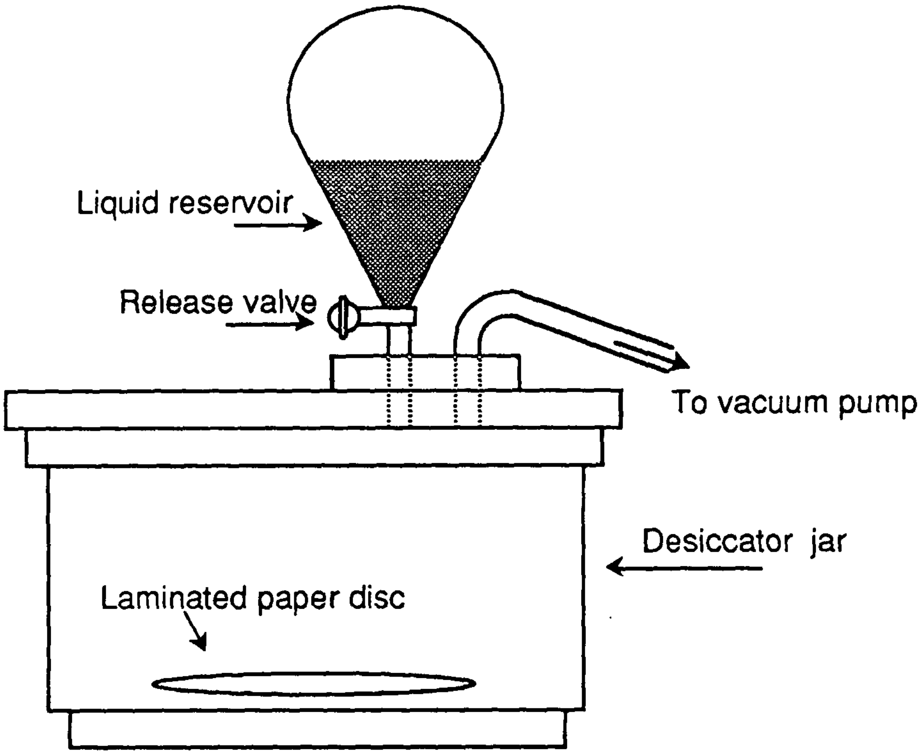


Figure 1. Test chamber used to induce in-plane flow in laminated paper disks.

As the fluid penetrates into the disk, photographs of the advancing liquid front are taken at intervals over a ten-minute period. The flat Lucite lid provides good optical access to the sample. The extent of liquid penetration at any time is related to the in-plane permeability components, as discussed below. Liquid boundary data from photographs are transferred to a Macintosh computer using a digitized tracing tablet and are then analyzed with a shape processing program, MacMeasure[9].

THEORY AND ANALYTICAL TOOLS

We desire to relate anisotropic permeability characteristics of a paper sample to the observed motion of a penetrating fluid boundary in an initially dry disk. Unfortunately, the problem of flow penetrating into a circular, anisotropic disk is not as simple as one might suppose. The analysis of this flow problem begins with Darcy's law:

$$\mathbf{v} = \frac{-\mathbf{K}\cdot\nabla P}{\mu}, \tag{1}$$

where $\mathbf{v}$ is the superficial velocity vector, $\mathbf{K}$ is a second-order permeability tensor (a 3x3 matrix with only three independent terms [10]), μ is the viscosity, and ∇P is the pressure gradient. For flow in the plane of paper, we can neglect the z-direction and choose an xy-coordinate system in which the permeability tensor becomes a 2x2 diagonal matrix:

$$\mathbf{K} = \begin{bmatrix} K_x & 0 \\ 0 & K_y \end{bmatrix}, \qquad (2)$$

where K_x and K_y are the in-plane permeability components in the x and y directions, respectively. In paper, these directions will generally correspond to the cross-direction (defined here as x) and the machine-direction (defined as y) [6]. The ratio of the y-component to the x-component is defined as α, the in-plane anisotropy ratio:

$$\alpha = \frac{K_y}{K_x} . \qquad (3)$$

If the two components are identical (α=1), the medium is said to be isotropic in the plane. Values of α other than 1 indicate anisotropy due to a pore structure with a preferred orientation in the plane. Under normal papermaking conditions, fibers (and thus pores) tend to be preferentially oriented in the machine direction, so α values greater than 1 are expected.

For the region of the paper which is saturated with the presumably incompressible liquid, the continuity equation is simply

$$\nabla \cdot \mathbf{v} = 0. \qquad (4)$$

Substitution of Equation 1 into Equation 4 results in:

$$\nabla \cdot \mathbf{K} \cdot \nabla P = 0. \qquad (5)$$

A solution to Equation 5 requires specification of the boundary conditions. For the case of interest, the fluid at the outer edge of the paper disk, where r = r_0, is at atmospheric pressure. The fluid at the inner edge of the penetration zone can be taken as the vapor pressure of the liquid, assuming complete evacuation of the test chamber before the fluid was added. The location of this inner boundary changes continually with time.

By neglecting pressure gradients and fluid flow in the θ-direction, the simplified continuity equation can then be integrated and combined with Darcy's law to obtain the following expression for the velocity of the fluid boundary at any time:

$$v_b(\theta) = \frac{dr_f(\theta)}{dt} = \frac{K(\theta)\,\Delta P}{\varepsilon\mu\, r_f(\theta)\,\ln[r_f(\theta)/r_o]}, \tag{6}$$

where v_b is the velocity of the fluid boundary at a particular value of θ, ε is the porosity, r_f is the corresponding radial location of the advancing fluid front, K is the effective permeability along a ray of constant θ, ΔP is the total pressure drop across the fluid zone of the disk, and r_o is the radius of the outer edge of the paper disk. The effective permeability at constant θ is given by:

$$K(\theta) = K_x \cos^2(\theta) + K_y \sin^2(\theta)\,. \tag{7}$$

Equation 6 can be rearranged and integrated, holding θ constant, to give:

$$\left(\frac{r_f(\theta)}{r_o}\right)^2 \left[2\ln\left(\frac{r_f(\theta)}{r_o}\right) - 1\right] + 1 = \frac{4K(\theta)\,\Delta P\, t}{\varepsilon\mu\, r_o^2} \tag{8}$$

from which the effective permeability in any direction can be obtained from an image of the fluid boundary at a known time, t. A value for α at any time t can then be obtained using values for r_f in the x- and y-directions:

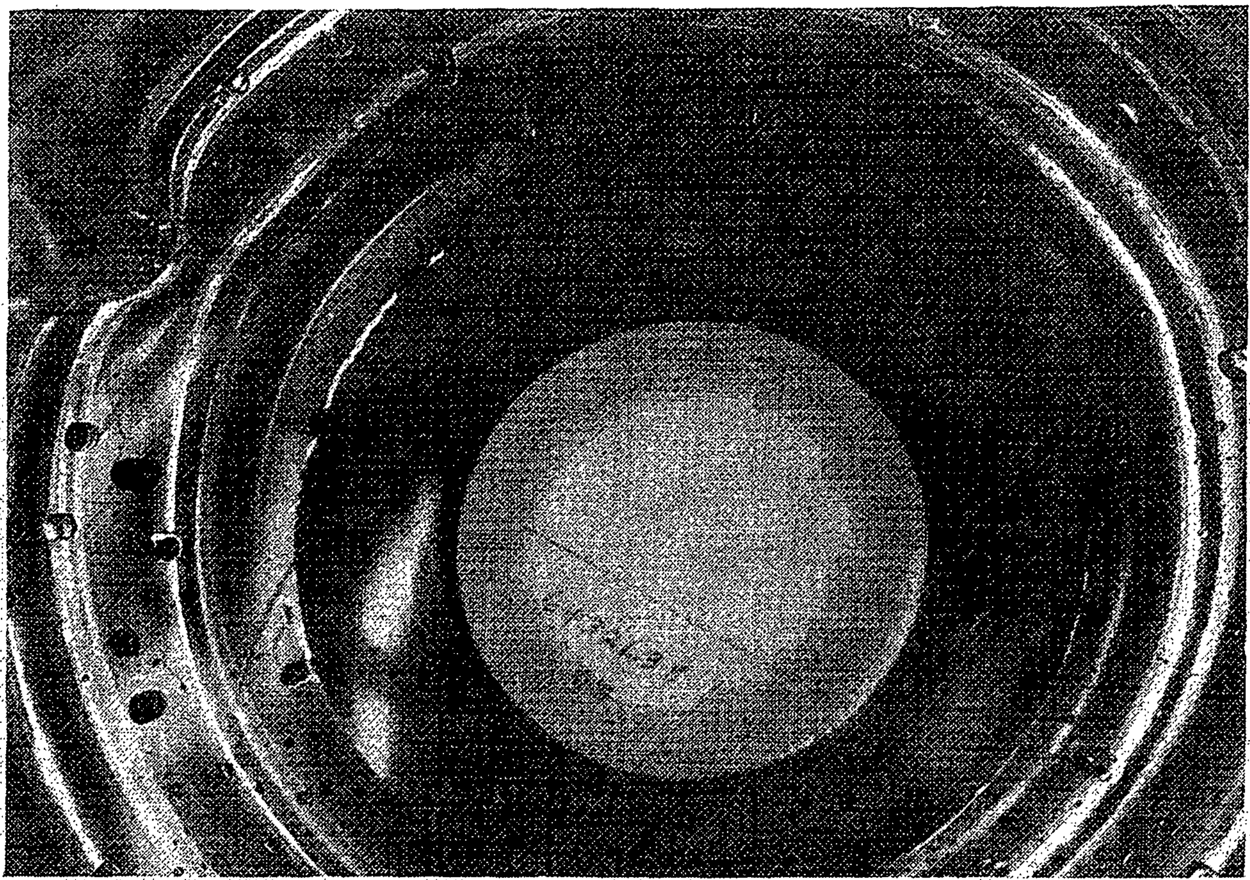
$$\alpha = \frac{K(\pi/2)}{K(0)} = \frac{r_f(\pi/2)^2\left[2\ln\left(\frac{r_f(\pi/2)}{r_o}\right) - 1\right] + 1}{r_f(0)^2\left[2\ln\left(\frac{r_f(0)}{r_o}\right) - 1\right] + 1}\,. \tag{9}$$

Equation 9 is an approximation based on the neglect of angular flow. The error introduced by this assumption approaches zero for α near unity, and, drawing from a numerical investigation of a related problem[6], should be less than about 5% for α values as high as 2.0

RESULTS

Figures 2a-2e are taken from a series of successive photographs of the fluid boundary as it moved into one of the laminated disks used in this study. The initially circular fluid boundary assumes a more elliptical shape as it moves into the disk of paper. The more rapid penetration in the machine direction (marked by a line on the sample) indicates that MD permeability (K_y) is greater than the CD permeability (K_x). Small instabilities on the fluid boundary grow in time, forming fingers which can become significant as the fluid nears the center of the disk.

a)



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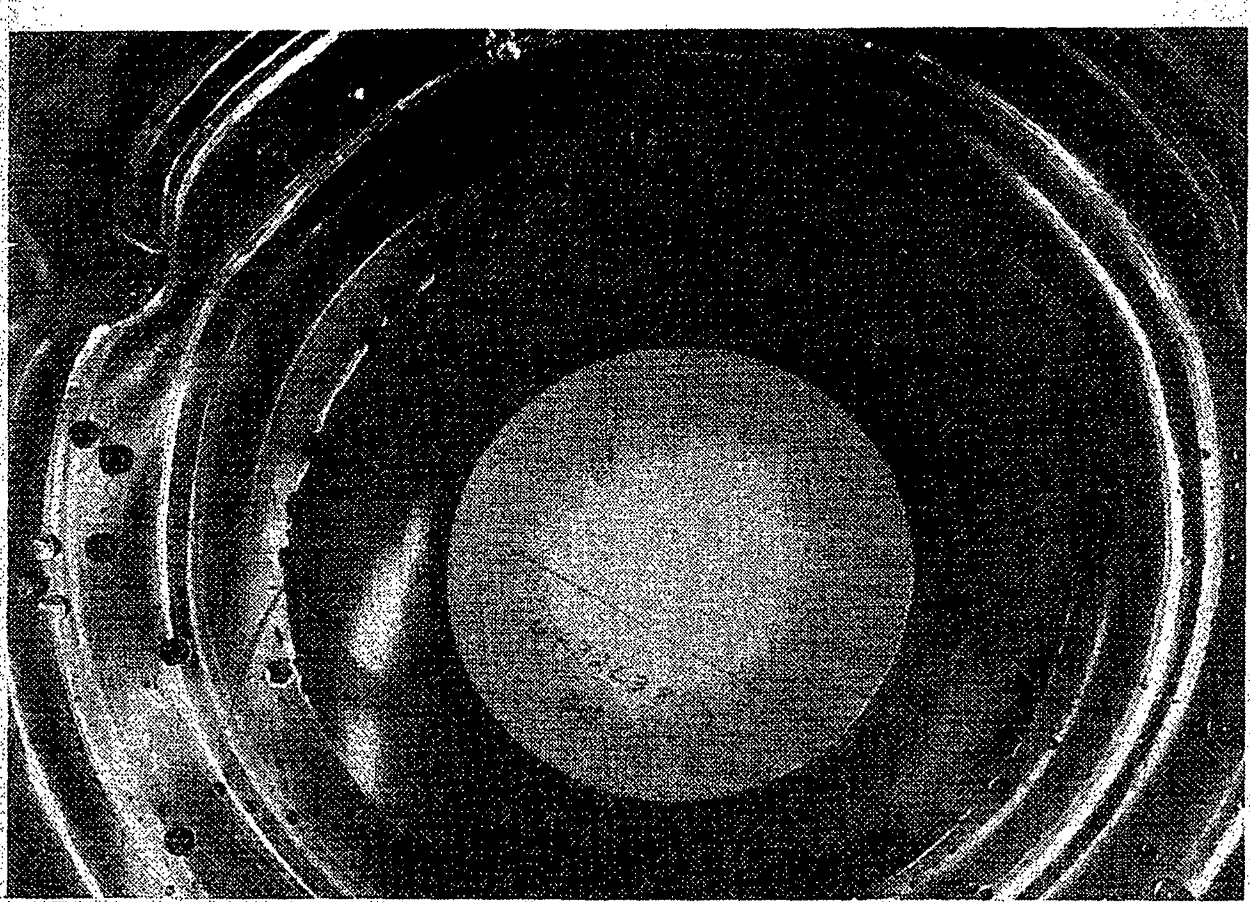

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a)

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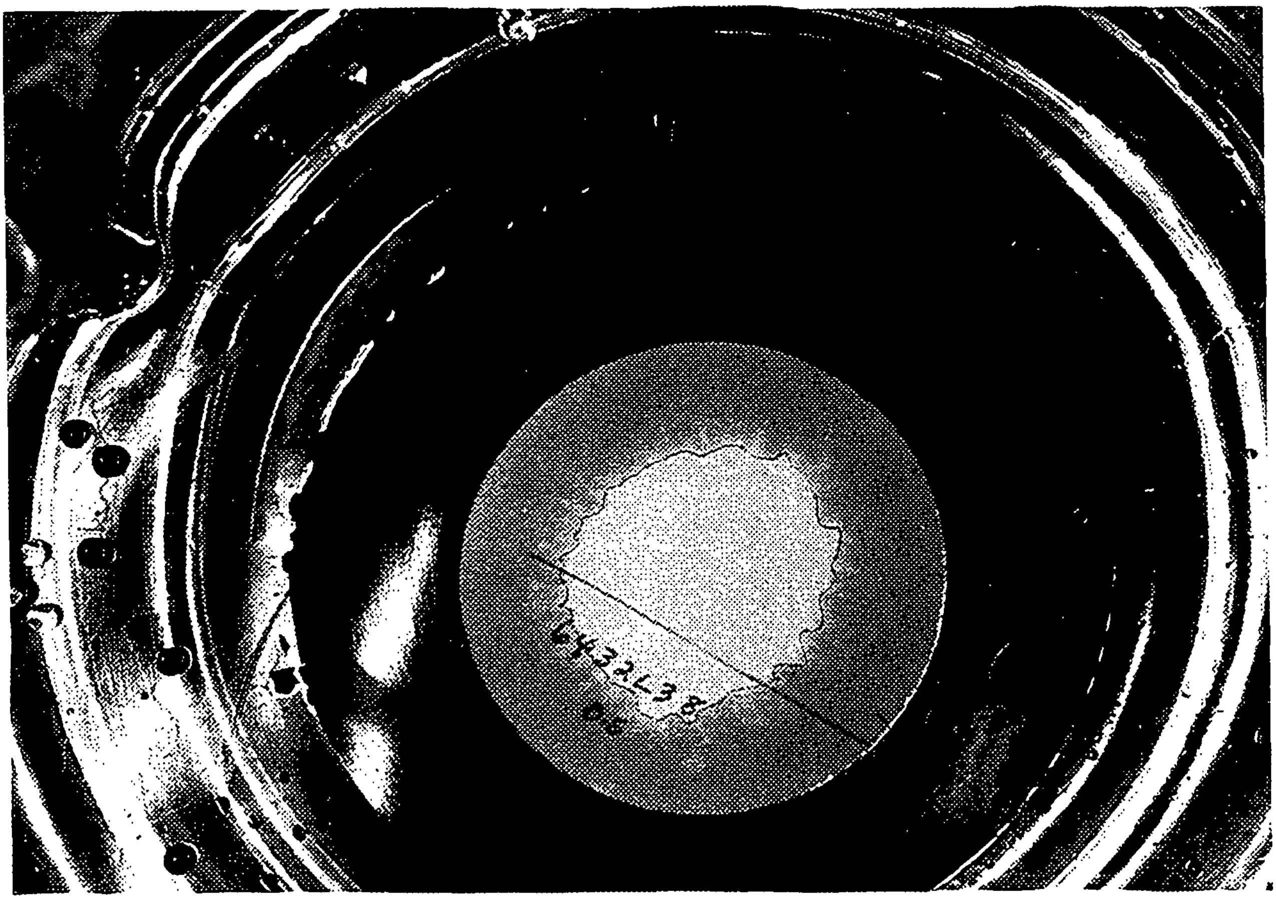

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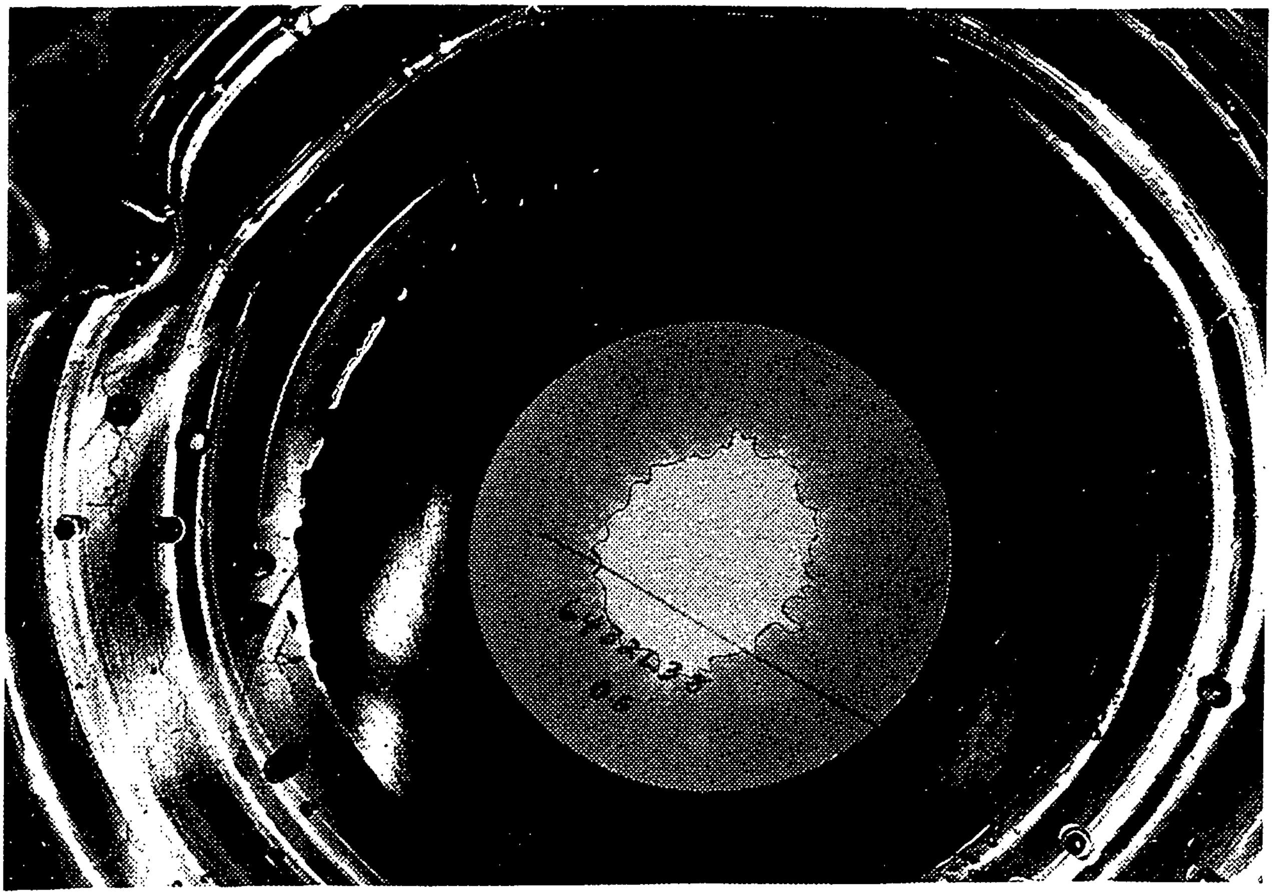

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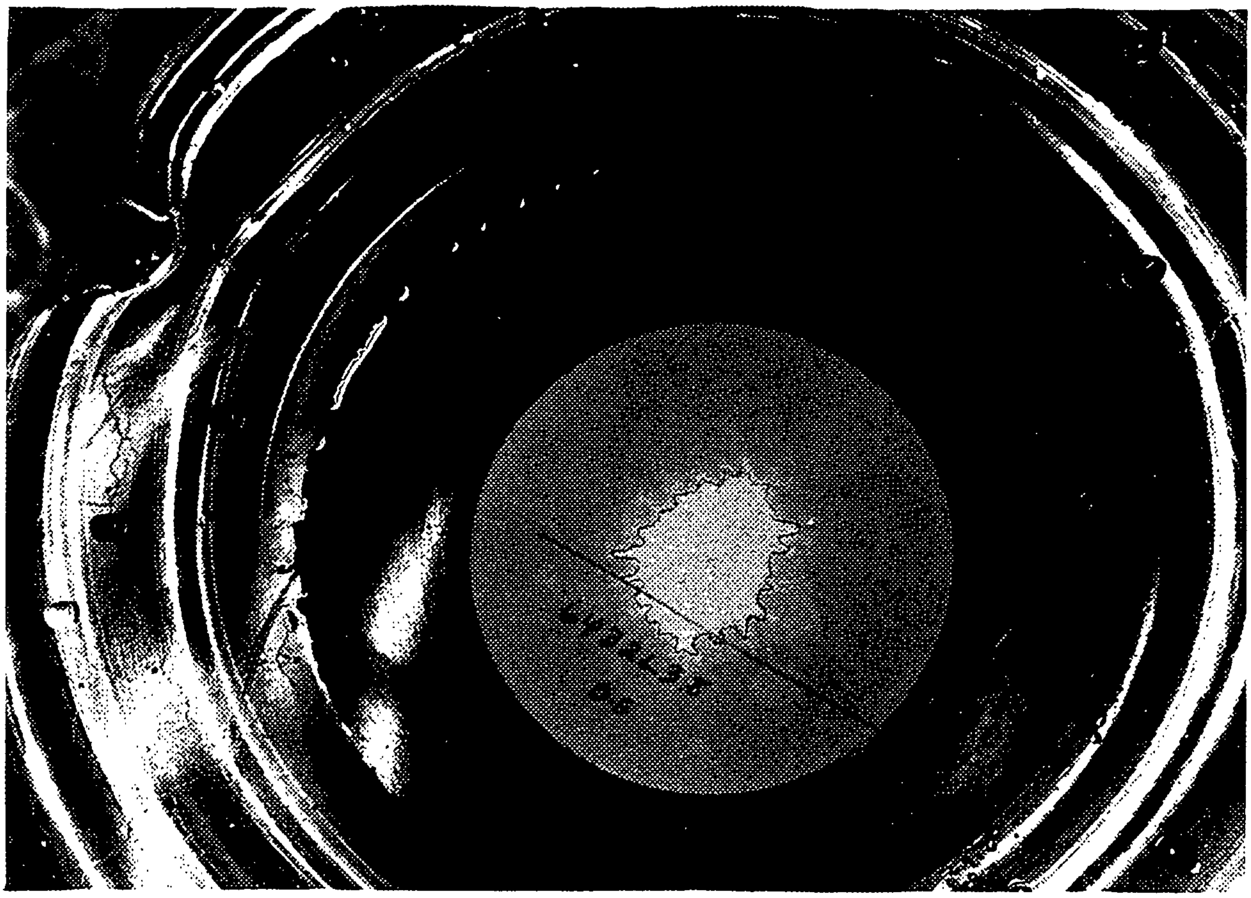
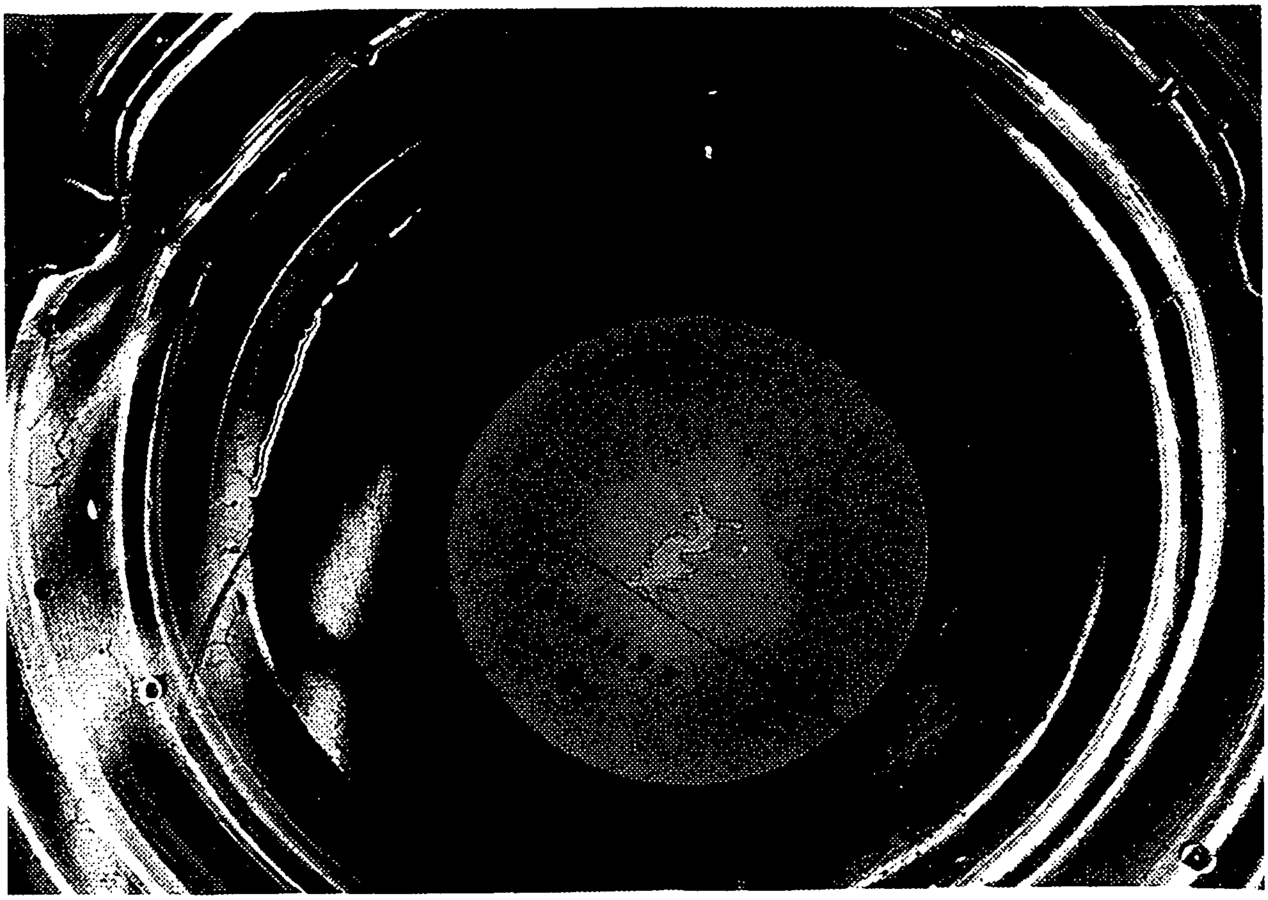


e)



Figures 2a-2e. Sequential photographs of the moving fluid boundary during an edge-penetration run in a laminated disk of paper. The machine direction is indicated by a line on the sample.

Values for α from five samples are shown in Figure 3. For the first several seconds, there is a large amount of scatter in the measurements of fluid boundary shape. Much of this error may be due to inherent fluctuations in the formation of the paper, which have a large effect on the initial flow but become averaged out once the fluid has penetrated further into the paper. After about 30 seconds, successive measurements of α become fairly stable, with a value of about 1.2 being typical. This means that for a given in-plane pressure gradient, fluid would flow about 20% faster in the machine direction than in the cross-direction.

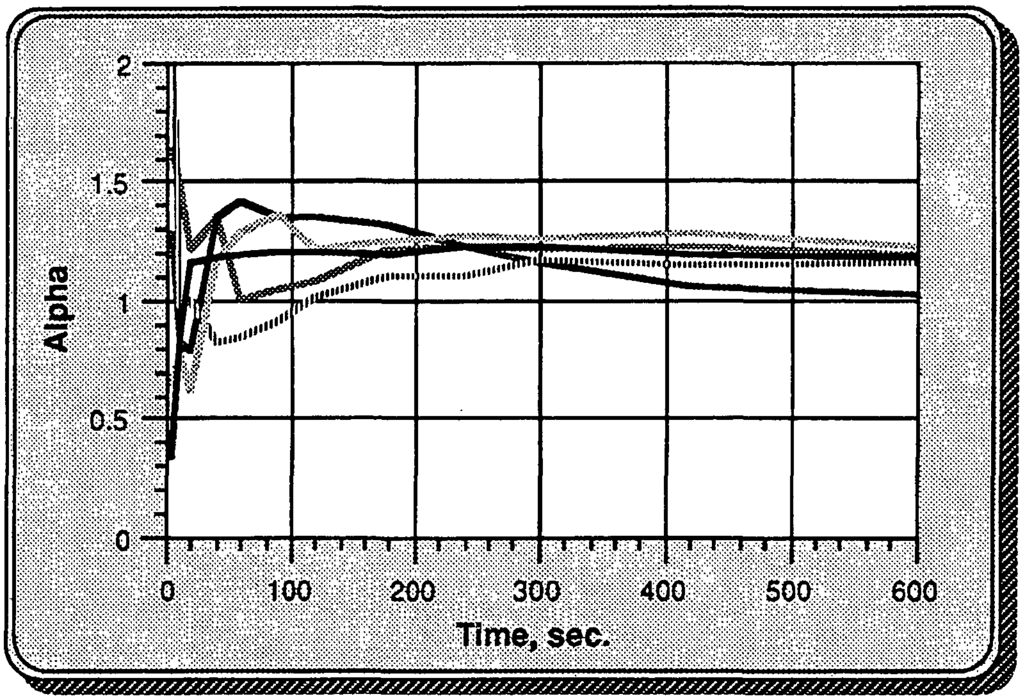


Figure 3. Measurements of α in time for five typical samples used in this study.

Values of average in-plane permeability can also be obtained at each measurement time. A convenient estimate of the average in-plane permeability can be obtained by using Equation 8 with an average r_f. The average r_f is defined as the radius of a circle having the same area as the dry zone of the disk encompassed by fluid boundary. The MacMeasure Software[10] package was used to evaluate the bounded areas from the photo series. Using this program simply consists of tracing the objects of interest on a digitized graphics tablet. The program then calculates the area of the tracing, the perimeter, and a shape factor. The major axis of the approximately elliptical fluid boundary was taken as the machine direction. A repeatability study was carried out on four replicated tracings of both the outer circle and the boundary of the noncircular wetting front.

The test showed that variability due to the method was less than one percent for the circles but close to six percent for the odd-shaped inner areas. The porosity, a needed parameter for permeability determination with Equation 8, was measured using mercury porosimetry.

Figure 4 shows the results obtained from the same set of measurements discussed above. Note that the apparent average in-plane permeability continues to decrease in time. This is apparently due to the effect of fiber swelling. As the fibers become wet, they begin to swell, leaving less open void space for liquid flow. Fibers near the edge of the disk, which have been exposed to liquid for the longest time, are expected to have swelled the most, and the freshly wetted fibers at the inner boundary of the fluid zone will be the least swelled. The apparent permeability is an average of the various permeability values along a ray from the outer edge toward the center, and will tend to be weighted toward the lower permeability values. The decrease in effective porosity with fiber swelling was not measured; only the initial porosity from mercury porosimetry measurements was used in Equation 8. This will affect the estimate of K but not of α.

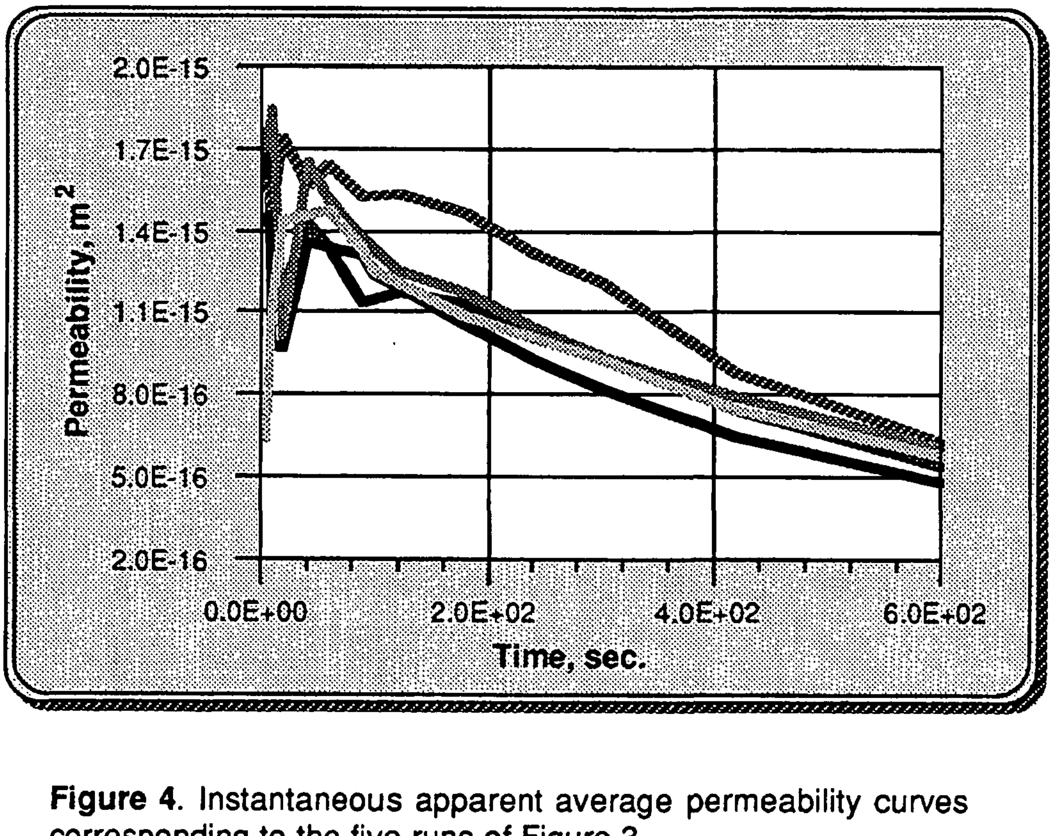


Figure 4. Instantaneous apparent average permeability curves corresponding to the five runs of Figure 3.

It is significant that the measured values of α remain fairly constant in time while the permeability continues to decrease. This implies that the degree of in-plane anisotropy, characterized by α, is not a function of the saturation or the degree of fiber swelling. The same observation was made by Lindsay[6], who found similar α distributions in initially dry and initially saturated blotter paper samples. Further experimentation is necessary to determine whether this observation is generally true.

DISCUSSION

Implications

The in-plane anisotropy of a fiber network is expected to be affected primarily by fiber orientation. Happel[10] and others[11,12] have shown that the flow resistance of liquid over a bank of parallel, aligned cylinders is less when the flow is parallel to the cylinders than when it is perpendicular. By analogy, if the fibers in a sheet of paper are preferentially oriented in the machine direction, then the MD permeability should be higher than the CD permeability. Likewise flow in the z-direction, for which most of the flow will be perpendicular to the fibers, should have the lowest permeability. Lindsay[6] found z-direction permeabilities in two types of paper to be lower than the average in-plane permeabilities; no z-direction measurements were made in the laminated disks of this study.

The observation that α does not change significantly after fiber swelling may indicate that fiber orientation, which should not change with swelling, is the dominant factor affecting α. Further experimental work is needed to determine general relationships among the components of the three-dimensional permeability tensor as a function of fiber orientation.

Limitations

The experimental work and the data reduction procedures rely on several simplifying assumptions which may introduce some error. For the α values seen in this study, the error due to the approximate data reduction method should be negligible. Uncertainties due to nonuniformities in the paper may have a larger effect. The presence of residual air in the laminated disk has also been neglected, although a significant back pressure may begin to build as the fluid boundary moves near the center of the sheet. Measurements of fluid boundary growth were made over a period of 10 minutes, which was not long enough for the fluid to penetrate all the way to the center of the disk.

Capillary forces were also neglected in this study, since the atmospheric driving force for the lateral penetration was expected to be greater than the capillary flow forces in the sample. This was confirmed by testing similar samples that differed only in the amount of internal sizing. While significantly different contact angles were measured due to the different sizing levels, no difference in penetration behavior was seen. This indicates that the flows of this study were not affected by surface tension effects. It should also be noted that disks which were not evacuated by vacuum took approximately 20 hours for the fluid to penetrate as far as it did in 10 minutes with the evacuated disks, again indicating that capillary flow was relatively negligible in the experiments reported here.

CONCLUSION

A simple experimental method has demonstrated the potential to provide information about in-plane anisotropy in paper permeability. To interpret the fluid boundary shape data from the experimental method, the flow problem of injection in a laminated, anisotropic disk had to be solved with an approximate method. This study supplements a technique for measuring in-plane permeabilities introduced in a previous paper[6]. In the photographic paper examined here, the machine direction permeability was found to be about 20% higher than the cross-direction permeability, consistent with expectations based on simple flow models, and consistent with related measurements in other papers using a different approach. In this study, it was also observed that the degree of in-plane anisotropy did not change significantly with fiber swelling.

The results of this and the previously reported study on anisotropic permeability are among the first such results to appear in the literature; much more work is needed to more fully understand the importance of in-plane flow in paper, and the relation of paper structure to anisotropic permeability.

ACKNOWLEDGMENT

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