A Simple Method for Calculation of the Permeability Coefficient of Porous Media

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ABSTRACT: The fluid storage capacity of porous media, such as paper and paperboard, is mainly determined by its porosity, whereas the absorption and spreading rate is determined by the permeability. A simple method for calculation of the permeability coefficient of porous media is described. The permeability coefficient may be calculated by Darcy's equation [1] using the Parker Print Surf porosity, which is primarily sensitive to air permeability [2-5]. The permeability coefficient may be used for ranking porous media in fluid absorption and spreading rate and for estimation of pore size. Likewise, the coating thickness required for given barrier and printing performance may be estimated.

Application: The permeability coefficient can be used as a quantitative tool to predict the barrier performance of porous media.

Porous media, such as paper and paperboard, contain small open spaces, voids and pores, distributed throughout solid matrices [6]. Adsorption and absorption of liquids through these pores have repercussions with respect to the barrier properties and to the ultimate strength of the sheet [7-9]. Moreover, the pores have an effect on the interaction between printing inks and paper [10-12].

Paper is laminated, surface sized or extruded with a variety of film formers and plastics to lower its permeability to moisture, water vapor, water and other solvents, and paper is internally sized to prevent excessive spreading of inks [13-15]. In addition, in many converting operations, where liquids are forced into the sheet by the action of a nip, the porous structure of the base sheet is important. Thus, the characterization of the porous property, i.e., permeability, is of major importance for predicting the barrier and printing properties of paper products.

PERMEABILITY AND POROSITY

Permeability is the most important physical property of a porous medium, while the porosity is its most important geometrical property. The permeability describes the conductivity of a porous medium with respect to fluid flow, whereas porosity is a measure of the fluid storage capacity of a porous material. Permeability describes how easily a fluid is able to move through the porous material. Thus, it is related to the connectedness of the void spaces and to the pore size of the paper. It is calculated using a formula widely known as Darcy's Law [1].

For quality control, units of measurement may be irrelevant, but for scientific research it is essential to be able to express results in absolute units [3]. Permeability measurements expressed in units of airflow or time cannot be compared directly with pore dimensions. For this reason, there is a need for calculating permeability results in appropriate units, which is area or square of length. As such, it can be interpreted in terms of an effective capillary cross sectional area or diameter [16].

DARCY'S LAW

Current equations describing fluid transport in porous media are based on semi-empirical equations derived in the 19th century by Darcy [1] for single- phase flow and in the 20th century for multi-phase flow. These equations describe the average behavior of a mixture of a porous medium and one or more fluids. Darcy's law describes the kinetics of fluid flow through porous media in terms of the driving force and the permeability of the medium.

Darcy's Law (1)

Where:

$$Q = \frac{K}{\eta}\frac{\Delta P}{\Delta L}A$$

Q = flow rate (m^3/s)

K = permeability coefficient, (m^2)

ΔP = pressure drop or difference, (Pa)

L = flow length or thickness of test sample, (m)

A = area of cross-sectional area to flow, (m^2)

η = fluid viscosity, (Pa-s)

The permeability coefficient K depends on the combination of the fluid and porous material used. The greater the value of K, the higher will be the rate of flow of a fluid through a material. While Darcy's equation was formulated from experimental data over a century ago, it was only recently proved theoretically.

Resin	Solids content, %	pH	Viscosity, cps	Avg. Particle Size, µm
MB-3640	54.5 - 55.5	6.5 - 7.5	400, max	250 - 325

Table I. The characteristic of the resin

Mineral Pigment	Aspect Ratio	Avg. Particle Size, µm	BET Surface Area, m^2/g
Nanoclay	200-400	0.12-0.14	
Kaolin clay # A	10-20	0.15-0.20	20-22
Kaolin clay # B	25-35	0.45-0.55	16-18
Kaolin clay # C	50-60	0.45-0.55	18-20

Table II. The characteristic of the mineral pigments

Substrate	Grammage, g/m^2	Internal Sizing	PPS Porosity, mL/min
Unbleached kraft base paper	70	None	2136
Solid Bleached Sulfate (SBS) baseboard	266	None	284

Table III. The characteristic of the base substrates

Mokadam [17] showed that Darcy's equation is a special case of a general equation that he derived for flow through porous media using irreversible thermodynamics. Therefore, Darcy's equation is a theoretically and experimentally valid law in the laminar flow for all porous media. The applicability of Darcy's law was also confirmed experimentally to porous media such as paper [18-21].

Ridgeway, et al. [11] used Darcy's equation to compare the liquid permeability coefficient with the Bendtsen air permeability coefficient. Their results showed close agreement with a few exceptions. Generally, we expect the liquid and gas permeabilities of a given medium to be different, but of the same order of magnitude. The gas permeability should be close to the liquid permeability of a perfectly wetting liquid. Differences in permeability for different wetting conditions are suggested by the Lucas-Washburn equation [21,22].

In this paper, Parker Print Surf (PPS) porosity measurement values (mL/min) are used to calculate the permeability coefficient of various papers, in order to predict the barrier resistance to gasses.

EXPERIMENTAL

This work is divided into two phases: (1) to develop a method for calculation of a permeability coefficient, and (2) to study the effect of pigment type, coating application methods, coating pickups and coat weight on measured permeability coefficients.

An acrylic polymer emulsion Lipacryl-MB3640 was used for size press and blade coatings. The viscosity, average particle size, solid content and pH of the emulsion, according to manufacturer, are given in **Table I**. Lipacryl-MB3640 has been used by other researchers [9]. A low molecular weight ethylated starch was also used for size press and impregnator coatings.

The characteristics of nanoclay and kaolin clays, according to manufacturer, are given in **Table II**. Nanoclay was used for size press and impregnator treatment [23,24]. Three different aspect ratio clays were used for size press and blade coating. Two commercial papers, unbleached kraft paper and SBS baseboard, were used as the substrate for the dispersion coating.

The base substrates characteristics are given in **Table III**. The unbleached kraft paper was size press and impregnator treated [23,24]. The bleached kraft board was size press treated and blade coated. The Impregnator coater and size press pickups and application conditions are given in **Table IV**.

A Messmer Instrument PPS Model 90 was employed to measure Parker Print Surf (PPS) porosity [2-5]. TAPPI Standard Method T-555 was used to measure PPS porosity. The PPS Porosity was calculated as the mean of 10 readings at different locations.

PROCEDURE

Cellulose nitrate model papers of different pore sizes were used for test development. The physical properties of the cellulose nitrate papers are shown in **Table V**. The papers were conditioned and then tested for PPS porosity and caliper. The PPS porosity was measured at a 1000 kPa clamping pressure. The standard parameters for the PPS tester used were as follows:

Fluid (air) viscosity (η): 1.80075E-05 Pa.s (Ns/m^2) at 23°C

Standard pressure drop (ΔP): 6.17 kPa [25,26]
Area of cross-section (A): 10 cm^2 [26]

By incorporating the standard pressure drop (ΔP), fluid viscosity (η) and cross-section area (A) values into equation (1), the following relationship was obtained:

PIGMENT LOADING (%)	IMPREGNATOR COATER			SIZE PRESS	
	Pond Pressure (PSI)	Shoe Pressure (PSI)	Pickup (%)	Solids (%)	Pickup (%)
0	5	40	10.4	22	14.1
0	10	40	13.5	14	9.5
0	15	40	14.3	6	5.6
3	5	40	7.8	22	14.6
3	10	40	10.0	14	10.3
3	15	40	12.2	6	5.9
3	20	40	14.0		
9	5	40	6.3	22	14.3
9	10	40	8.0	14	9.2
9	15	40	11.1	6	6.4
9	20	40	12.9		
9	25	40	15.5		

Table IV. Impregnator Coater and Size Press Pickups and Application Conditions

$$\text{Permeability Coefficient, } K\ (\mu\text{m}^2) = 0.048838{*}Q\ (\text{ml/min}){*}\ L\ (\text{m}) \tag{2}$$

An example calculation of the permeability coefficient for 0.20 µm pore size model paper using equation (2) follows:

Parker Print Surf flow rate (Q) at
1000 kPa: 477 mL/min

Thickness of 0.20 µm model paper
(L): 124 µm

Permeability coefficient, K =
0.00289 μm^2

Additional examples for cellulose nitrate papers are shown in Table V. Note that the permeability coefficients reach an asymptotic value as a function of the thickness of the paper sample, while the PPS porosity values are inversely proportional to thickness. As a result, the PPS porosity will continuously decrease, where as the permeability coefficient will reach an asymptotic value as a function of the thickness of the sample. This property is evident from **Fig. 1**.

COATINGS FOR SBS

Six different coatings were prepared with pigments of different aspect ratios and resins. The coatings varied in the amount of dry pigment added on dry weight of resin. The amount and aspect ratio of pigment was varied to determine the influence on the porosity and permeability coefficient of the SBS paperboard in comparison to a conventional pigmented coating. The pre-dispersed clays at 62% solids and a synthetic resin at 54% solid were used for all coatings. The pigment was blended into the appropriate amount of resin to yield the desired coating suspension. The coating solids and viscosity were measured.

All size press coatings were applied at 30% solids and room temperature. The viscosities of the coatings were all below 150 mPas. All blade coatings were applied at 58% solids and room temperature. The viscosities of the coatings were all below 400 mPas. The coatings were applied to a 266 g/m^2 SBS baseboard using a laboratory size press and cylindrical laboratory coater. The porosity and permeability coefficient of the samples were measured. The influence of pigment aspect ratio on permeability coefficient was studied.

COATINGS FOR UNBLEACHED KRAFT

Three different coatings were prepared with a low molecular weight ethylated starch. The coatings varied in the amount of dry pigment added on dry weight of starch (0%, 3%, 9%). The clay was added to determine its influence on the porosity and perme-

Sample I.D.	Avg. Pore Size (µm)	Avg. Thickness (µm)	PPS Porosity (ml/min)		Permeability Coefficient (μm^2)	
			Avg.	Std.	Avg.	Std.
Cellulose Nitrate-Low Porosity	0.20	124	477	5.05	0.00289	0.0000310
		270	245	3.21	0.00323	0.0000429
		381	164	2.33	0.00304	0.0000439
Cellulose Nitrate-Medium Porosity	0.45	124	787	7.59	0.00476	0.0000466
		270	403	6.89	0.00531	0.0000921
		381	273	3.68	0.00508	0.0000694
Cellulose Nitrate-High Porosity	0.80	98	2614	18.40	0.01251	0.0000892
		229	1382	12.25	0.01546	0.0001388
		321	932	9.74	0.01461	0.0001547

Table V. Permeability coefficients of model papers

ability coefficient of the unbleached kraft paper in comparison to starch alone. The clay was pre-dispersed at 15% solids using a high shear Cowles disperser. An anionic dispersant was added at a rate of 1% on weight of pigment (dry/dry). The viscosity of the pigment dispersion was 99 mPas (Brookfield #3 spindle, 60 rpm, 38°C). The elevated temperature of the clay was the result of the high shear energy used to disperse the pigment.

The starch was jet cooked at 24% solids and the viscosity measured at 27°C. The viscosity of the starch was 55 mPas @ 60 rpm. The pigment was then blended into the appropriate amount of starch solution to yield the desired sizing solution. The solids in the coatings were measured using a Labwave solids analyzer, and the Brookfield viscosities were measured at 60 rpm with an LVT digital viscometer. All coatings were applied at approximately 20% solids at 32°C. The viscosities of the coatings were all below 100 mPas.

The coatings were also applied to an unsized, 70 g/m² unbleached Kraft base paper using an impregnator coater [23,24]. Because the pickup on the impregnator could be controlled by increasing the reservoir pressure, the coating solids did not have to be adjusted and remained at 20% for all runs.

Adjusting the reservoir pressure also enabled the depth of penetration to be controlled [23,24]. At the higher pigment loadings, more pressure was need-

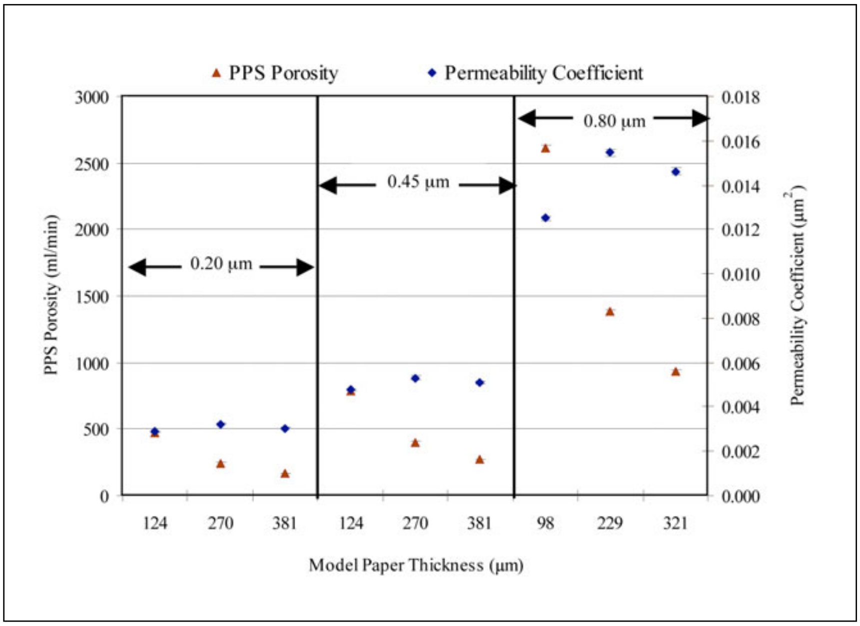

Figure 1. Comparison of permeability coefficients and PPS porosities of model papers

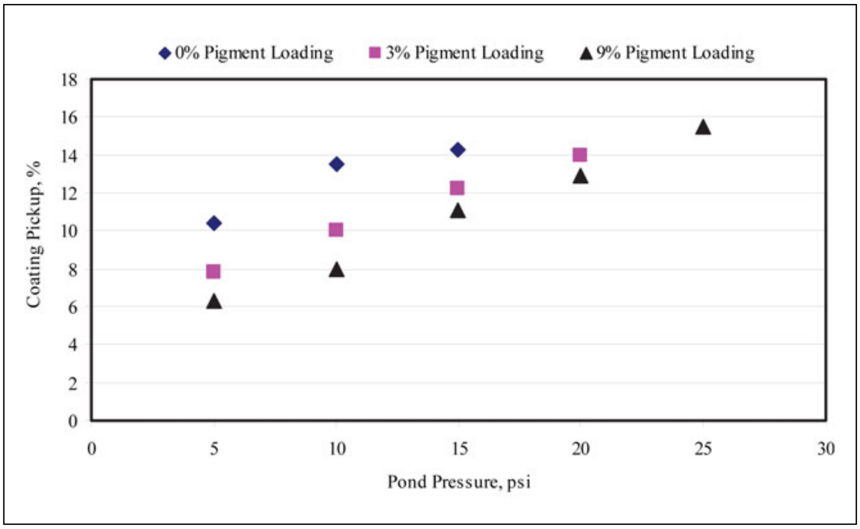

Figure 2. Influence of impregnator pond pressure on coating pickups

Pigments	Formulation (wt %)	Coating Method	Avg. Coat Wt. (g/m²)	PPS Porosity (mL/min)		Permeability Coefficient (µm²)	
				Avg.	Std.	Avg.	Std.
#A	50% Pigment + 50 % Resin	Size Press	8.7	13.7	1.53	2.03E-04	2.26E-05
#B			8.3	8.8	0.84	1.25E-04	1.20E-05
#C			8.2	10.0	1.18	1.45E-04	1.71E-05
#A	91% Pigment + 9.0% Resin	CLC-Blade	9.3	11.3	1.18	1.64E-04	1.71E-05
#B			8.5	12.1	0.95	1.72E-04	1.35E-05
#C			10.2	10.5	1.35	1.52E-04	1.95E-05

Table VI. Influence of pigments aspect ratio on PPS porosity and permeability coefficient

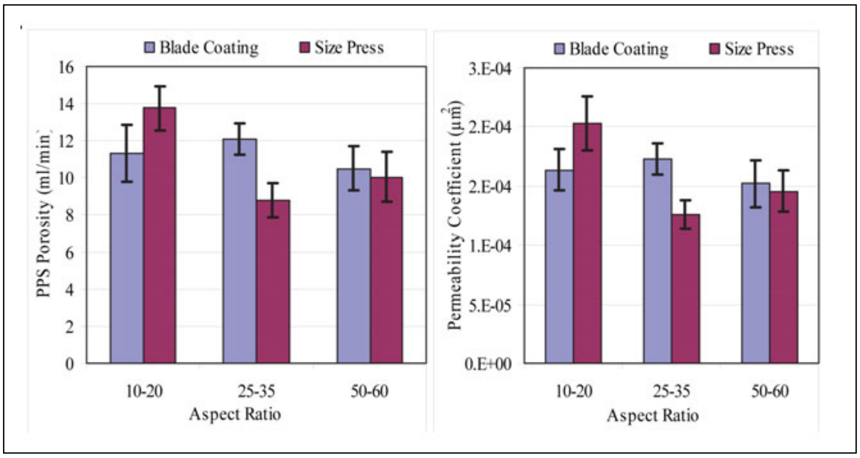

Figure 3. Influence of pigments aspect ratio on PPS porosity and permeability coefficient

ed to drive the coating into the sheet. The reservoir pressures were gradually increased until evidence of the coating passing through to the backside of the sheet was seen. Evidence of coating penetration completely through the sheet was confirmed by the change in color of the backside of the sheet after drying.

To control the pickup at the size press, the solids of the coatings were adjusted down by adding dilution water. The pickups obtained for each coating application are shown in **Table VI**. The pickup at the size press increased linearly with coating solids for all pigment levels. The influence of impregnator reservoir pressure on pickup is also shown in **Fig. 2**. For all levels of pigment applied, the pickup increased with increasing reservoir pressure. As the level of pigment increased, more pressure was needed to push the coating into and through the base-sheet, indicating a packing influence from the pigments.

RESULTS FOR SBS

The PPS porosity and permeability coefficient values are given in Table VI for the different pigments and coating conditions. The permeability coefficient was calculated from PPS porosity results, as described above. The dependence of PPS porosity and permeability on aspect ratio of the pigments is shown in Table VI and **Fig. 3**. Size press treated samples gave lower PPS porosity and permeability than the blade-coated samples for the medium aspect ratio pigments.

The low and high aspect ratio showed little, if any, dependence on application method, probably because both methods are in the low shear regime for the low aspect ratio pigments and both are in the high shear regime for the high aspect ratio pigments. Thus, there was no apparent dependence on the different shear rates for the two methods. The high aspect ratio pigment produced the lowest PPS porosity and permeability for the blade coated samples, but not significantly, since that case also had the highest coat weight. PPS porosity and permeability values showed a difference

Pigment (%)	Impregnator Coater					Size Press				
	Pickup (%)	PPS Porosity (mL/min)		Permeability Coefficient (µm^2)		Pickup (%)	PPS Porosity (mL/min)		Permeability Coefficient (µm^2)	
		Avg.	Std.	Avg.	Std.		Avg.	Std.	Avg.	Std.
0	10.4	547.5	148.7	0.0040	0.0011	5.6	1446.6	151.3	0.0093	0.0010
0	13.5	485.6	56.4	0.0037	0.0004	9.5	1322.1	141.4	0.0088	0.0009
0	14.3	447.2	45.7	0.0035	0.0004	14.1	1270.9	168.4	0.0085	0.0011
3	7.8	468.4	56.2	0.0035	0.0004	5.9	1445.8	189.5	0.0097	0.0013
3	10.0	390.0	58.8	0.0029	0.0004	10.3	1193.3	146.7	0.0081	0.0010
3	12.2	320.7	32.1	0.0024	0.0002	14.6	1031.9	133.8	0.0070	0.0009
3	14.0	287.7	39.4	0.0023	0.0003					
9	6.3	384.0	29.8	0.0028	0.0002	6.4	1249.6	130.2	0.0081	0.0008
9	8.0	239.8	37.5	0.0017	0.0003	9.2	1020.6	109.7	0.0068	0.0007
9	11.1	177.5	29.9	0.0013	0.0002	14.3	856.4	74.9	0.0057	0.0005
9	12.9	131.2	10.5	0.0010	0.0001					

Table VII. Influence of saturation treatment on PPS porosity and permeability coefficient of different samples

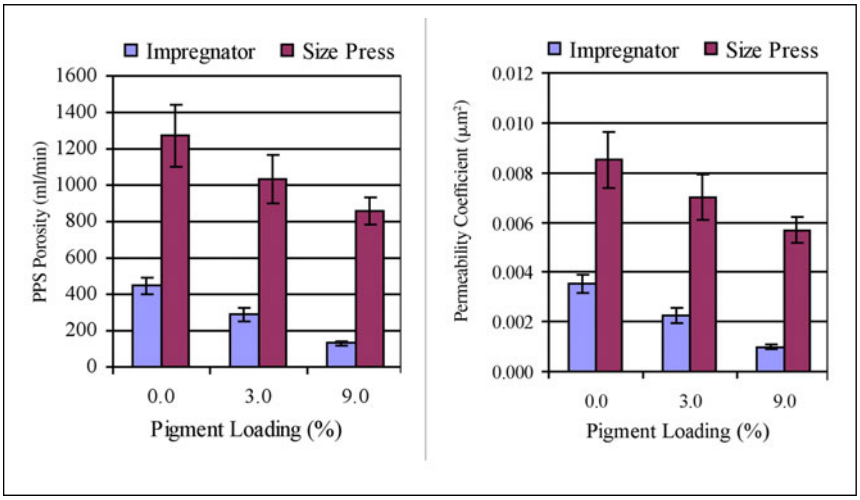

Figure 4. Influence of pigment loading on PPS porosity and permeability coefficient of size press and impregnator treated papers

between the two application methods at all pigment-loading levels.

The aspect ratio of the pigments was found to have a more pronounced effect on the permeability of the size press coated samples then the blade coated samples. The low permeability of the size press coated samples resulted in a greater resistance to moisture vapor transmission under high humidity and temperature conditions. These results will be presented in a future publication. At this time, we have accounted for total thickness, however work to account for the contribution of coating thickness to the effective permeability of the coated paper are in progress.

RESULTS FOR UNBLEACHED KRAFT

The PPS porosity and permeability coefficient values are given in **Table VII** for different conditions (base paper- PPS porosity = 2136 mL/min, permeability = 0.01476 µm^2). **Fig. 4** shows the influence of pigment loading (14% pickup, dry basis) on sheet porosity and permeability.

Impregnator treated samples gave lower porosity and permeability than size press. The significantly lower porosities of the impregnated treated samples indicate that this method is better for reducing the permeability of the papers or closing off the sheet. PPS porosity and permeability values showed a significant difference between the two application methods at all pigment-loading levels. The addition of nanoclay significantly reduced the porosity of both the impregnated and size press treated papers.

CONCLUSIONS

We have developed a simple method for calculating the permeability coefficient of gasses through porous media. The proposed permeability formula, based on Darcy's law, shows promise as a quantitative tool to predict the barrier performance of porous media. We have demonstrated its application to porous media, using different grades of coated and uncoated paper across a wide range of PPS porosities. The permeability coefficient, as calculated here, will reach an asymptotic value as a function of thickness of the sample, whereas the PPS porosity will continuously decrease.

For a uniform medium, the asymptotic permeability will be observed at very small thicknesses and corresponds to the permeability of the uniform medium. The asymptotic permeability for a laminated porous medium (e.g., coated paper) will be determined by the permeability of the flow rate controlling layer, (e.g., the coating). Use of this calculation tool provides more useful information to the papermaker, because it provides insight into the contribution of thickness and interconnectivity of the pores of the samples in comparison to airflow measurements. We found that the coating application method had a significant effect on the permeability coefficient. The permeability varied with coating pickup, pigment loading, and depth of penetration. **TJ**

Submitted: November 28, 2005
Revised: April 12, 2006
Accepted: April 12, 2006

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INSIGHTS FROM THE AUTHORS

In quality control testing, permeability measurements of paper have been expressed in units of airflow or time, which cannot be compared directly with pore dimensions. For this reason, we wanted to present permeability results in appropriate units, which is area or square of length. This way, permeability can be interpreted in terms of an effective capillary cross sectional area or diameter.

We wanted to develop a standard test for the measurement of barrier performance of porous media. The permeability coefficient could then be used for ranking porous media in fluid absorption and spreading rate and for estimation of pore size. Likewise, the coating thickness required for given barrier and printing performance could also be estimated.

The most difficult aspect of this research was finding the standard parameters for the PPS tester and model substrate for development of the permeability coefficient equation. We were able to find PPS tester standard parameters from literature and TMI personnel.

Now that we have developed a correlation between permeability coefficient and barrier performance of porous media, we hope this calculation tool will be used to provide more useful information to the papermaker. This information will give insight into the contribution of thickness and interconnectivity of the pores of the samples in comparison to airflow measurements.

At this time, we have accounted for total thickness. However, work to account for the contribution of coating thickness to the effective permeability of the coated paper is in progress.

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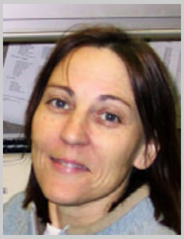
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