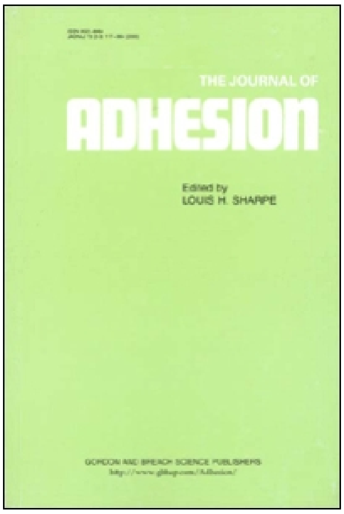

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The Peeling Behavior of Pressure Sensitive Adhesives from Uncoated Papers

Robert Pelton[a]; Wei Chen[a]; Hong Li[a]; Michael R. Engel[a]
[a] McMaster Centre for Pulp and Paper Research Department of Chemical Engineering, McMaster University, Canada

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THE PEELING BEHAVIOR OF PRESSURE SENSITIVE ADHESIVES FROM UNCOATED PAPERS

Robert Pelton
Wei Chen
Hong Li
Michael R. Engel
McMaster Centre for Pulp and Paper Research
Department of Chemical Engineering
McMaster University, Canada

When a tape based on pressure sensitive adhesive (PSA) is peeled from paper, either the tape comes off leaving a clean paper surface or the paper undergoes catastrophic cohesive failure which is the delamination of fiber layers in the paper sheet. The objectives of this work were to determine the links between paper properties and peel characteristics. Peel tests and microscopic analysis of a variety of handmade and commercial papers yielded the following conclusions. The tendency for paper failure is not very sensitive to surface energy. Paper roughness and density seems to be the dominant factors. A relatively smooth but weak paper will give a clean peel whereas a strong but rough handsheet or filter paper will always give paper failure. Lamination pressure is also important. Peel force increases with lamination pressure up to a limit where paper failure begins and the peel force plummets. The peel response of common paper types is mapped onto a 2-D surface whose axis reflects paper surface chemical and structural properties. It is proposed that the initiation of paper failure in peel occurs at fiber ends.

Keywords: PSA paper interactions; Peeling from paper; Paper failure; Release paper; Peel mechanics; Paper failure criteria

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Michael R. Engel is also affiliated with 3M company, 3M Center, St. Paul, MN 55144-1000.

Address correspondence to Robert Pelton, McMaster Centre for Pulp and Paper Research, Department of Chemical Engineering, McMaster University, Hamilton, Ontario, Canada L8S 4L7. E-mail: peltonrh@mcmaster.cn

INTRODUCTION

Paper remains one of the highest-performing, cost-effective packaging materials in part because it is a nearly prefect substrate for pressure sensitive adhesives (PSA) and tapes. Compared with common plastic films, paper has a rough, high energy surface which strongly interacts with inks and adhesives. Thus, paper sheets can be glued or taped to give exotic, engineered shapes required for modern packaging.

The manufacture of non-packaging paper grades also depends in part upon the favorable interactions between adhesives and paper. PSA-based tapes fulfill the crucial role of splicing together paper rolls. This is a particularly challenging application. Adhesion must be instantaneous when forming a flying splice and the resulting joint must be strong enough to survive printing or converting. In recent years the requirement for repulpability has been a further challenge for PSA developers [1]. Splices, address labels and other PSA-containing products often cause troublesome deposits when present in recycled pulp. Modern tapes with more hydrophilic adhesives can dissolve or be dispersed in the repulper to give fewer deposition problems.

PSA-based tapes are also an important tool for paper science. All z-directional paper strength tests involve fixing paper to testing equipment with PSA [2]. The key assumption is that the adhesive does not influence paper strength.

The commercial importance of PSA technologies has led to hundreds of patents and scientific papers addressing the physics and chemistry of PSAs and the major results are summarized in recent texts [3, 4]. However, in spite of all this work, only three scientific papers have focused on the interactions of PSA tapes with paper [5, 6, 7]. We suspect that this lack of scientific activity is because, in many cases, when testing PSA formulations with paper, it is the paper that fails giving limited information to the PSA formulator. By contrast, PSA peel tests from clean metal surfaces give cohesive failure of the tape and the corresponding peel force is sensitive to the PSA composition. The key findings from the literature are now summarized.

Bikerman treated paper as a porous medium and discussed the flow of PSA into the paper structure [5, 8]. He also was the first to report that peeling across a cut edge of paper greatly enhanced the chances of initiating paper delamination. Note that most types of papers consist of a layered structure of non-interwoven fibers, thus cohesive failure appears as a crude delamination.

Yamauchi and coworkers reported the first systematic peel studies from paper [6, 7]. Key observations were that measured peel force increased with peel rate until the onset of paper delamination after which the peel force was lower. They also showed that smooth, machine-finished papers were less prone to delamination than were handsheets or other rough papers.

Paper delamination in peel has been used to estimate the z-direction (*i.e.*, normal to the surface) strength of paper [2, 9, 10]. In a series of papers Yelon and coworkers analyzed the mechanics of paper bending and peel delamination [11, 12, 13]. They proposed measuring delamination force as a function of the minimum radius of curvature of the paper [12]. The peel work extrapolated to zero radius of curvature is a measure of the true delamination work.

Finally, this subject is unique because perhaps every reader has inadvertently conducted many experiments themselves in an effort to remove tape from paper without ripping the paper. Experience has taught us that to minimize paper failure we must peel the tape as slowly as possible. Experience also shows that recently applied tape is generally easiest to remove.

In this paper we present results of a scientific investigation of PSA/paper interactions with an emphasis on identifying the criteria under which paper fails. This extends the pioneering work of Yamauchi and coworkers by including the effects of lamination pressure, paper surface energy and peeling direction. Also a new approach to classifying papers with respect to peel behavior is outlined and ideas are presented regarding the initiation of paper delamination.

EXPERIMENTAL

Materials

A series of commercial and a second set of model pressure sensitive adhesives were employed in this work. The commercial tapes were 3M tape No. 405, No. 411 and No. 9954. 3M No. 405 is a repulpable splicing tape. It consists of green acrylic adhesive layers coated on both sides of a bleached tissue carrier and with a pink film liner. 3M No. 411 is a Flexomount^{TM} printing tape, with adhesive layers coated on both sides of a gray vinyl carrier and with a kraft liner. The thickness of adhesive layer is 0.139 mm and the thickness of vinyl carrier and paper liner are both 0.102 mm. 3M No. 9954 is another repulpable double-coated splicing tape, with green acrylic adhesive layers coated on a bleached tissue carrier, all supported on a white release paper.

Three model adhesives based on copolymers of acrylic acid (AA) and butyl acrylate (BA) were kindly provided by 3M Canada. The molar ratios of AA to BA used to prepare the adhesives were 10/90 in adhesive 924, 6/94 in adhesive 921 and 2/98 in adhesive 918. The

TABLE 1 The Designation and Description of Commercial Papers Used in This Work

Designation	Description
Copy Paper 1	Photocopy paper supplied by Canon Canada Inc. Basis weight 75 g/m^2. Item No. 9-20015-D1. (88521 1193R)
Copy Paper 2	Photocopy paper supplied by Domtar Canada Inc. Domtar® Windsor Copy Multipurpose Paper. Basis weight 75 g/m^2, acid free
SBKP	Unfilled semi-bleached kraft liner from Wausau Mosinee Corporation
Glassine	Staedtler-Mars Limited No. 946 1117S 100% rag.
Filter Paper	Whatman International Ltd. Maidstone, England. Cat. No. 1001 185, 185 mm

model adhesives were supplied as both 20% solutions and as tapes with coating weights of 43 g/m^2.

A variety of commercial machine-made papers was employed. The paper designations and descriptions are given in Table 1. Note that glassine is a general term used to describe paper which is dense and nearly transparent. Glassine is used for drawing, tracing and grease-proof packaging.

Handsheet Preparation

Handsheets are small, laboratory-prepared, paper sheets made by the filtration of a dilute pulp suspension using industry standard procedures (TAPPI T205 sp-95). In this work a softwood bleached kraft pulp provided by Avenor Corp. (now Bowater Canada), Thunderbay, Ontario, was used. Initial experiments revealed that handsheets prepared from untreated pulp were too weak to give useful peeling results. Mechanical treatment (beating) of the fibers is the standard approach to improved paper cohesive strength. In this work a Waring® CB10 blender was employed for pulp treatment and the details are described elsewhere [14].

Wet handsheets with a dry basis weight (mass/area) of 60 g/m^2 were constrained, to prevent wrinkling, by TAPPI standard rings and dried at room temperature. The handsheets were designated *Btt* where *tt* is the blending time in minutes. Some handsheet physical properties are given in Table 2. The cumulative pore volumes of the handsheets were measured by mercury intrusion at the University of Western Ontario.

TABLE 2 Mechanical Properties of Handsheets and Copy Paper. Peel Tests were Conducted at 180 Degrees at a Crosshead Velocity of 50 mm/min

Sample	Tensile index[a] Nm/g	Scott bond (kJ/m^2)	Surface roughness (μm)	Failure mode in peel (PSA 921 & PSA 924)
B0	25.4 + 2.4			paper
B20	77.1 + 5.3			paper
B40	85.2 + 3.6	190 ± 11	4.85 ± 0.63	paper
B40 300 psi*		199 ± 13	4.57 ± 0.37	paper
B40 700 psi*		19 ± 6	4.43 ± 0.40	paper
B60	83.3 + 10.3			paper
B90	81.0 + 5.0			paper
Copy Paper 1		214 ± 15	2.77 ± 0.22	
Copy Paper 2			2.03 ± 0.22 MD 2.18 ● 0.24 CD	

*hot pressed at 138 C for three minutes (300 psi = 2.07 MPa; 700 psi = 4.83 MPa).

[a] Tensile strength per width divided by the basis weight (mass per superficial area of paper).

Hot Pressing of the Handsheets

In an effort to modify paper surface structure, dry handsheets B40 were hot pressed at 138°C under pressure of 300 psi (2.07 MPa) or 700 psi (4.83 MPa) for 3 minutes using a Carver press. Paper physical properties including caliper and Scott Bond Strength were measured. The surface roughness was determined by a stylus profilometer (Mitutoyo® Surfest-211). Roughness is described by *Ra* which is the arithmetic mean of the absolute departures of the roughness profile from the mean [14].

Peel Test Sample Preparation

Samples for 180° peel tests employed three different tapes in a complex structure illustrated in Figure 1. The test paper was mounted onto a 2″ × 6″ (5.1 × 15.2 cm) stainless steel panel with 3M 411 double-sided mounting tape. A half-inch (1.3 cm) strip of 3M 405 tape was then applied across the end of the paper corresponding to the starting location for the peel test (see Figure 1). This "separation tape" was employed so that the leading edge of the paper could not induce paper failure. Bikerman and Whitney many years ago showed that peeling from or along a paper edge greatly increased the probability of initiating paper delamination [8]. The PSA test tape, which was to be peeled, was then applied to the paper surface. The tape width was less

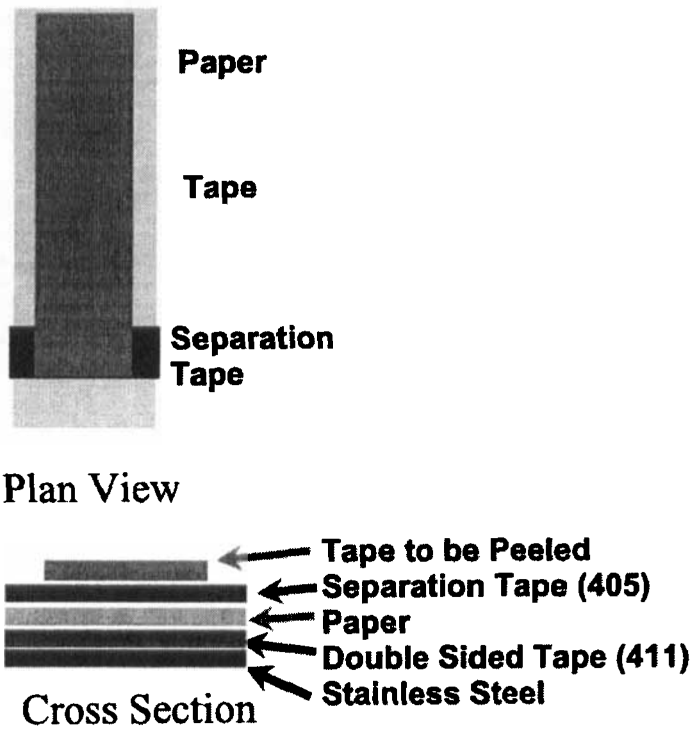

FIGURE 1 The construction of 180° peel samples.

than that of the paper to avoid edge effects. In cases where the test tape was double-sided, a SBKP paper was used as a top layer and was placed in the top clamp of the test machine.

In the final step, pressure was applied to the sample to laminate the PSA test tape to the paper. For most experiments a 4.5 lb (2 kg) rubber coated roller was hand rolled over the sample twice at a speed of about 5 mm/s. To study the effects of lamination pressure, samples were pressed with 4.5 lb (2 kg) and 15 lb (6.8 kg) rubber coated rollers on an automated single roller with a two passes at a speed of 305 mm/min. A range of pressures was obtained by varying the width of the PSA test tape. Peel tests were conducted within three minutes of lamination.

Peel Testing

Two instruments were employed for peel testing. An Instron® Model 1130 tensile tester with 500N load cell was used to perform a 180 degree peel test according to ASTM D3330-96. In this method, 18 cm × 2 cm tape strips were placed on the paper and pressed with a 4.5 lb (2 kg) roller twice at a velocity of approximately 5 mm/s. The 5 cm tape tab was fixed to a strip of SBKP paper (see Table 1). The end of the SBKP paper was gripped with the upper clamp while the panel with the paper was placed in the lower clamp. The typical crosshead velocity used was 50 mm/s. Experiments with this instrument were conducted under ambient conditions.

The second peel instrument was an Instron Corporation Series IX Automated Material tester located in a constant temperature (23°C) and humidity (50% RH) room. This instrument could be fitted with a horizontal slide for doing 90° peel tests.

ESEM Peeling

Peeling behavior was observed at high magnification with a Philips ElectroScan Environmental Scanning Electron Microscope System 2020 Version 3.2 fitted with a tensile stage. Special panels were prepared with a dimension of 5 mm × 40 mm × 1.5 mm. The peel angle was 180° and the peel rate was 10 µm/s.

Vapor Phase Sizing

Vapor phase sizing of the handsheet was performed in order to modify the paper surface energy while maintaining the paper structure. Following Swanson and Cordingly's method [15], 250 mg stearic acid (Sigma®, 99%) was dissolved in 25 mL toluene (10 mg/mL). One milliliter of the solution was added to a nearly horizontal 25 × 200 mm test tube and the toluene was allowed to evaporate at room temperature. The test tube was attached to a smooth glass surface placed on a level table by a thin double-sided adhesive tape to insure its horizontal position. Care was taken to ensure that the stearic acid crystals that formed with solvent evaporation were distributed uniformly along a line from the back to about 25 mm from the front of the tube. The tube was then heated at 105°C for 0.5 hr to remove residual toluene and to fuse stearic acid to the tube. The tube was then closed with a Teflon-covered rubber stopper and equilibrated in a 105°C oven for 0.5 hr. Handsheets, cut to 2.5 cm × 15 cm, were placed in the middle of the test tubes containing stearic acid for at least 16 h.

Fluorochemical Treatment of Handsheet

Following Dickson's [16] procedure, handsheets B40 were dipped into a 3% w/w fluorocarbon emulsion (3 M ScotchBan$^{\text{TM}}$ FC-845 Paper

Protector) in ethanol. Excess fluorocarbon solution was removed by pressing the handsheets between blotter papers. The handsheets were air dried and then cured by placing the sheets in a 105°C oven for 15 minutes.

Contact Angle Measurement

Contact angles were determined with a Ramé-Hart Model 100 contact angle goniometer. Paper samples were cut to 60 × 25 mm and fixed on microscopic glass sides by double-sided tape. A water drop (distilled followed by Millipore® Q treatment) with a volume of $4\,\mu L \pm 0.4\,\mu L$ was placed on the surface. The contact angle was determined by estimating the tangent normal to the drop at the intersection between the sessile drop and the surface 5–20 s after the application of the drop. The tests were done at room temperature and the humidity was not controlled. All the reported values were an average of at least 10 measurements.

RESULTS

The Behavior of Paper in Peel

Paper/PSA interactions were probed with peel tests. In the following sections results are presented as functions of the important variables. Three types of failure were observed—paper failure, interfacial failure and tape failure. Figure 2 shows three peel tape surfaces after peeling from paper. Paper failure is obvious and unequivocal; at least one layer of fibers is embedded in the tape after peeling. We use term “interfacial” to denote the situation illustrated in Case (a) where the tape appears to peel cleanly from the paper. However, microscopic examination of the tape surfaces after peeling often reveals small paper debris on the tape. We have used fluorescent labeled PSA to reveal microscopic PSA remnants on the paper after peeling. Thus, we believe that true interfacial failure rarely occurs in paper.

The middle picture in Figure 2 corresponds to mixed failure—initially the failure mode is interfacial until the moving tape front initiates paper failure. In cases where the paper and tape have the same width, paper failure often starts at an edge.

The peel force traces are characteristic of the failure mode as is illustrated Figure 3. In the case of paper failure, the maximum force occurs initially and it appears to represent the force required to initiate paper failure–we call the maximum the “Peak Force”. The corresponding “Mean Force” is low and the trace is noisy in the case of

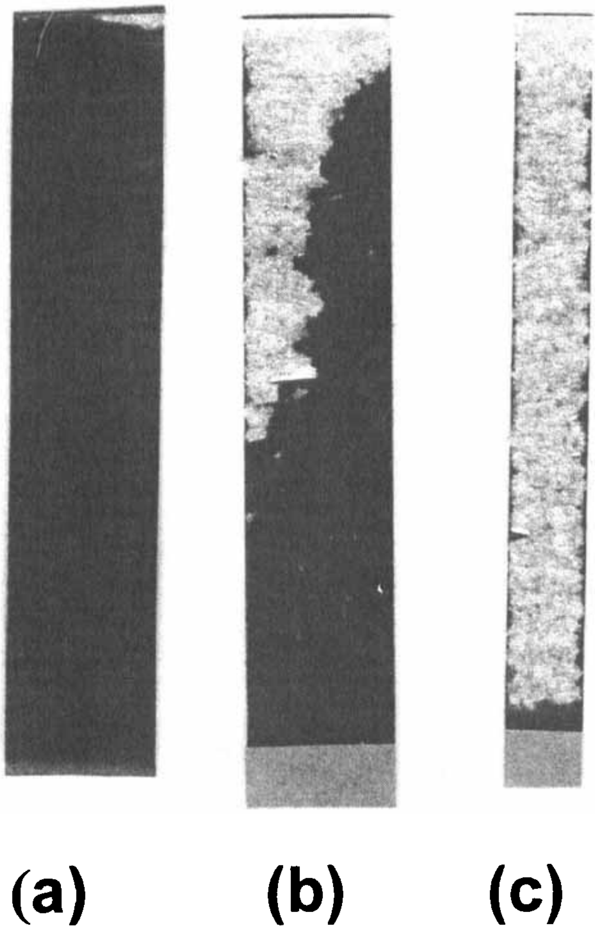

FIGURE 2 Black tape surfaces after peeling from white paper. (a) "Interfacial" failure, (b) mixed failure, and (c) is paper failure.

paper failure. By contrast, interfacial failure gives higher and smoother peel traces where the peak force is close to the mean force. Mixed failure generates an erratic peel trace.

Handsheets

Beating and Pressing

Handsheets (laboratory-made paper) were prepared from softwood kraft pulps by standard methods. Pulps were beaten with a blender

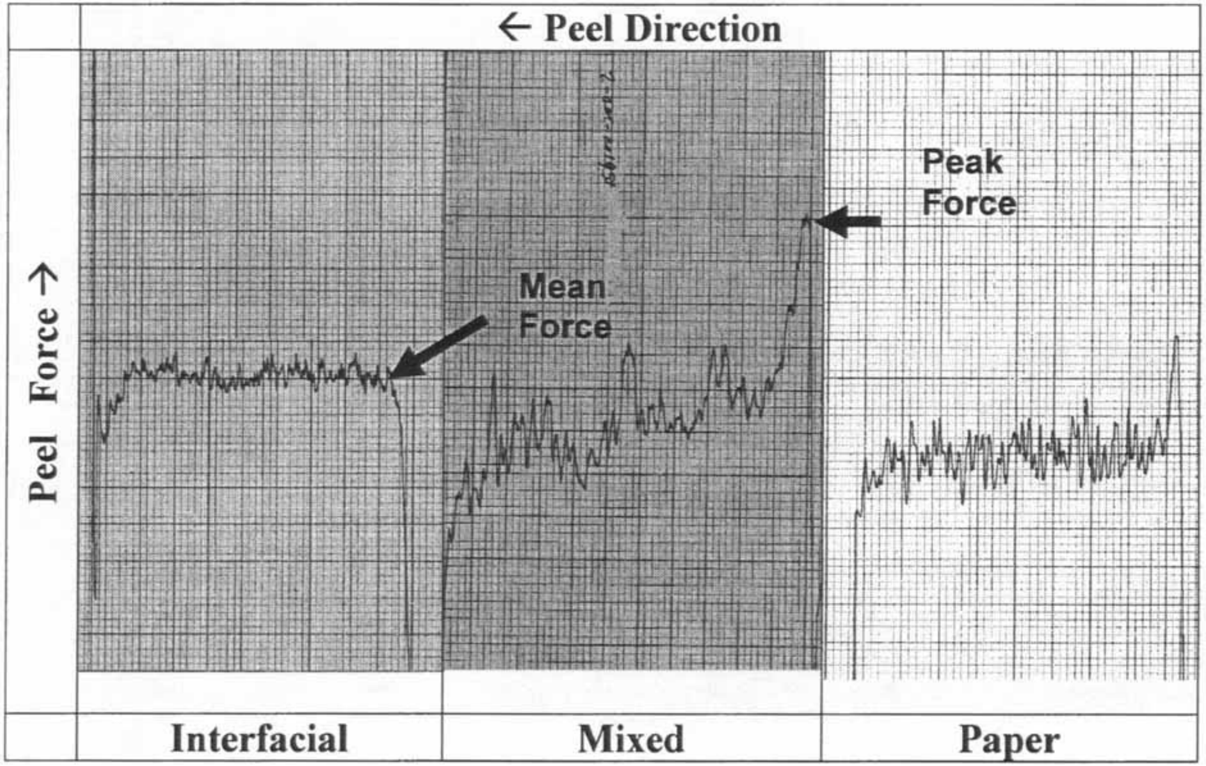

FIGURE 3 Examples of three types of peel traces. Peak Force (PF) is the force corresponding to the initiation of delamination whereas Mean Force (MF) is a measure of the Z-direction strength of paper. The vertical axis is peel force and the horizontal axis is the distance of the peel front from the beginning position from left to right. The sample width is 25.4 mm.

and peel measurements were made as functions of beating and pressing. Handsheet properties and the corresponding failure modes in peel are summarized in Table 2. B20 refers to handsheets prepared with pulp beaten in a blender for 20 minutes. For comparison, some properties of copy papers are shown. In every case reported, tape peeling from untreated bleached softwood kraft pulp handsheets resulted in immediate paper failure. By contrast, commercial xerographic papers can show interfacial or paper failure depending upon the details of the peel test.

Most of the handsheet tensile and Scott Bond strength parameters were comparable. On the other hand, the xerographic papers were smoother and often gave interfacial failure.

Surface Treatment

Bleached kraft pulp handsheets were sized by stearic acid vapors using the method of Swanson and Cordingly [15] in an effort to lower the surface energy of the paper. Table 3 gives the water contact angle as a function of exposure time to the stearic acid vapor. Although contact angles measured on rough paper surfaces are known to have dubious fundamental value [17], the results indicate the papers were fairly hydrophobic and that vapor contact time had a bigger effect on contact angle than beating. Results for two untreated commercial papers are also shown for comparison. Water drops rapidly penetrated the copy paper so measurements were a short time after drop application.

TABLE 3 The Water Contact Angles and Failure Mode of Handsheets as Functions of Treatment Time with Stearic Acid. Also Shown are Corresponding Properties of Untreated Commercial Papers and Handsheets Treated with Fluorochemical

Sample	Treatment time (h)	Contact angle (deg)	SD	Failure mode in peel
B60	40	114	3.1	paper
B60	16	87	3.8	paper
B90	40	103	4.6	paper
B90	16	89	3.8	paper
B40	16	95	5.1	paper
B40	40	111	2.2	paper
Copy Paper 1	0	85	3.5	
SBKP	0	112	3.4	
B40	Fluorochemical	122	6.7	Interfacial

Every handsheet treated with stearic acid displayed paper failure in peel. By contrast, the fluorochemical treated handsheets did give interfacial peel.

Commercial Papers

Machine-made papers tend to be smoother and the fibers more oriented than conventional handsheets. The following results show that machine-made papers have a much higher tendency towards interfacial failure than do handsheets.

Peel Rate, Model Adhesives

In a typical example of peel force *versus* peel rate for peeling from a strong flat substrate, the peel force increases until reaching a plateau value. Peeling from paper gives more complicated behavior. Figure 4 shows the mean peel force as a function of peel rate for three model, PSA coated tapes. SBKP is a semi-bleached kraft pulp liner and the PSAs are based on simple copolymers of n-butylacrylate and acrylic acid giving a range of properties. PSA924 caused paper failure at all peel rates and the peel force was insensitive to peel rate. PSA924 had the highest acrylic acid content. Acid groups are known to contribute to both the PSA cohesive strength and to the PSA adhesion with the paper [18].

PSA918, an experimental adhesive, was weak and gave cohesive failure at low peel rates. When the peel rate was fast enough to initiate interfacial failure, the steady state peel force dropped, presumably because the interface failed before energy was consumed by bulk adhesive flow.

Table 4 compares peel results for two types of machine-made paper. Copy Paper 1 gave higher peel forces with no paper failure under these conditions. A sample of Copy Paper 1 was delaminated by peeling with a very strong tape a high peel rate. A peel experiment was then conducted from the rough delaminated surface and only paper failure was observed. This emphasizes that surface structure is an important determinant of the failure mechanism on peel.

Lamination Pressure

Everyday experience has taught us that the tape adheres more strongly to paper if one presses on it during application. Furthermore, it seems reasonable to propose that the role of lamination pressure is to promote the flow of the adhesive into the paper structure. A series of peel experiments was conducted with 411 tape and Copy Paper 1. Before peeling, the tape and paper sandwich was pulled through a

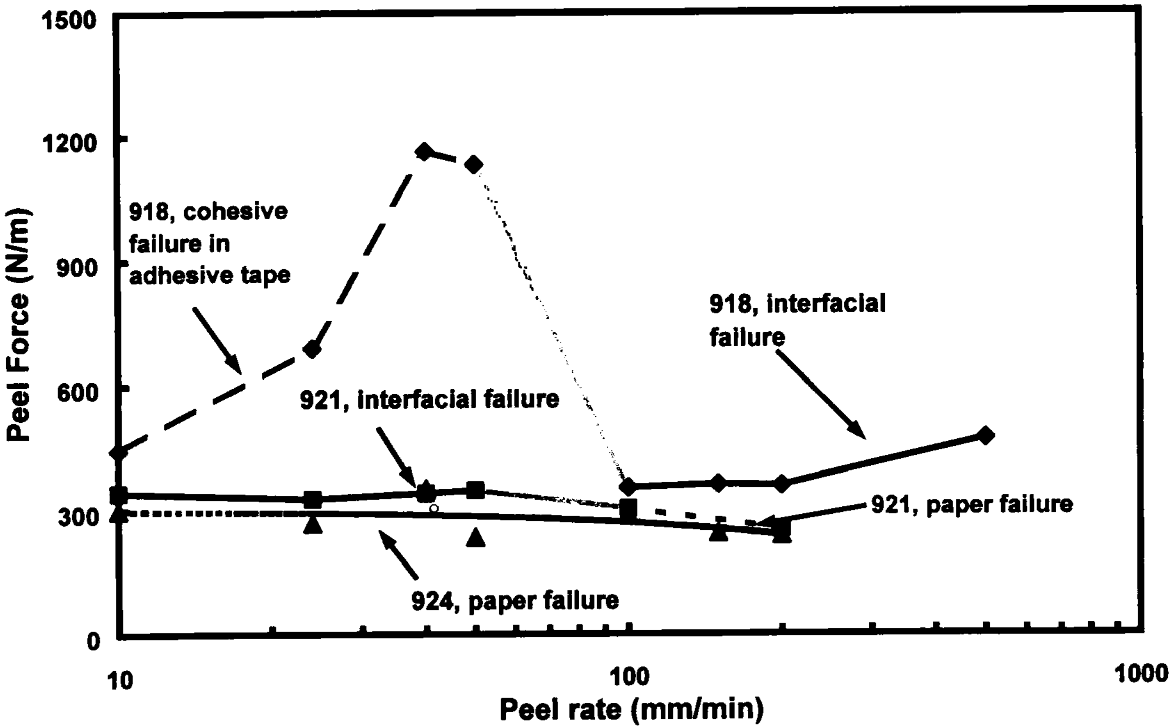

FIGURE 4 Peel force (180°) *versus* peel rate for model PSAs from SBKP.

TABLE 4 A Comparison of Peel Results for Two Commercial Papers. The Peel Rates and Angles were 50 mm/s and 180°

Paper	Tape	Mean peel force N/m	Failure mode
SKBP	PSA921	347	Interfacial with fiber pullout.
SKBP	PSA924	234	Paper failure
Copy Paper 1	PSA921	601	Interfacial
Copy Paper 1	PSA924	650	Interfacial
Delaminated Copy Paper 1	PSA921	–	Paper failure

rubber-coated calendar at a speed of 305 mm/min. The resulting peel forces are shown as functions of loading on the rollers in Figure 5. At the lowest peel rate, the peel force increased with lamination pressure for the first three samples after which delamination occurred (open points). Note that peak delamination forces (see Figure 3 for definitions of peak and mean forces) are shown. However, once delamination has initiated, the forces are low and not very dependent on peeling conditions. Thus, for the 50 mm/min data a little lamination pressures helps, whereas too much pressure leads to paper failure and a weaker structure in peel. As before, higher peel rates increase the probability of paper failure.

Peel Direction

Paper is made as a long continuous sheet by the filtration of a dilute wood pulp suspension through a plastic screen moving as a belt at high speeds. Because of the nature of this process, paper is inherently anisotropic. For example, the tensile strength measured when force is applied to the paper sheet along the direction in which the papermachine was running (*i.e.*, the "machine direction" (MD)) usually differs from the tensile strength measured at right angles in the cross direction (CD). This effect occurs because the fibers are preferentially oriented in the machine direction. In the case of peeling, the situation is even more complicated. When considering the machine direction (MD), there are actually two possible directions – towards the dry end and towards the wet end. It has been long known in the paper physics community that the failure mode in tape peel can be very different "towards the wet end" *versus* "towards the dry end". For example, Niskanen's book [19] presents Skowronski's data showing that the force required to delaminate paper is direction dependent. Surprisingly, there are no published results showing peel characteristics as a function of peel direction on paper.

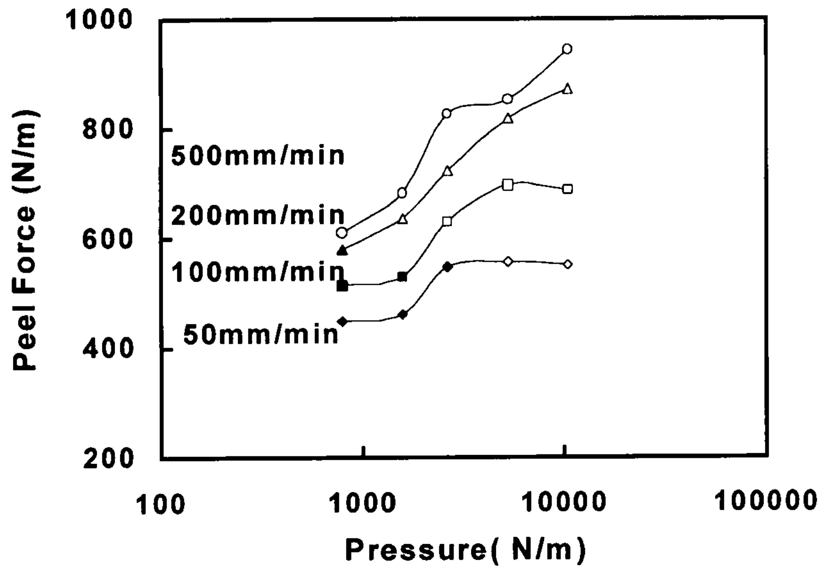

FIGURE 5 Peak peel force (180 degrees) *versus* lamination load for peeling 411 tape from Copy Paper 1. The solid points indicate interfacial failure and the forces shown are mean values, whereas the open points show paper failure and the forces shown are peak forces. The corresponding steady-state paper delamination forces are less.

Figure 6 shows the average peel force as a function of lamination pressure and peel direction. 3M No. 9954 splicing tape and Copy Paper 2 were used for this group of experiments. To obtain the copy paper sample, it was necessary to collect a cut and wrapped pack of paper in the mill so that the downstream direction could be assigned. At the two lowest lamination pressures the results were independent of direction, whereas, at higher lamination pressures, peeling towards the wet-end gave higher peak forces and paper failure and peeling towards the dry-end gave lower forces.

SEM – The Initiation of Paper Failure

Tape and paper samples were routinely examined by SEM after peeling. Furthermore, 180-peel experiments were conducted in a micro-tensile stage mounted in the chamber of an environmental scanning electron microscope (ESEM). Figure 7 shows a tape sample after peeling in conditions giving interfacial failure. The PSA surface shows mainly indentations corresponding to the location of embedded fibers before peel. However, a few fibers picked from the paper are visible.

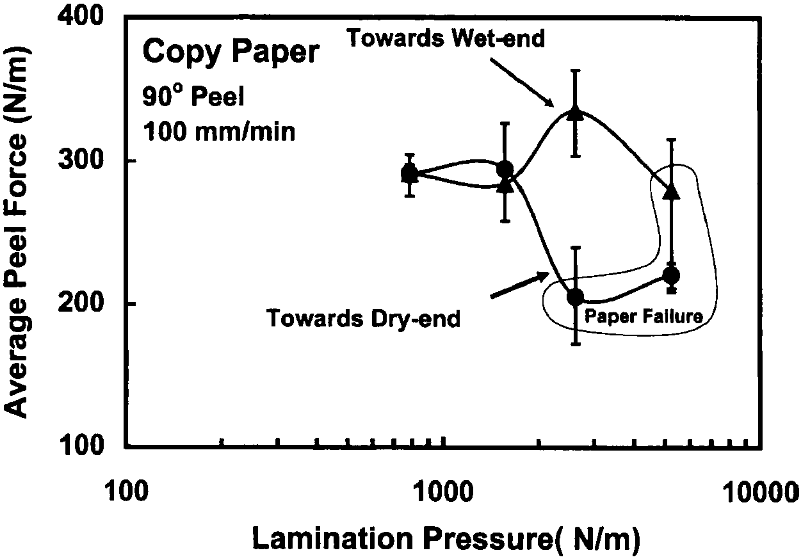

FIGURE 6 The influence of peeling direction on peel force and lamination pressure on Copy Paper 2.

The largest fiber in the upper right hand quadrant appears to have one end embedded in the PSA and there is a PSA tether stretching from the adhesive surface to the fiber surface, much like a guy wire.

In mixed failure mode experiments it is possible to view the region on the tape corresponding to the onset of paper failure—Figure 8 shows an example. The transformation from interfacial (right) to paper failure (left) occurred abruptly over a distance scale of about two fiber diameters.

Figure 9 shows a frame captured from the video sequence of peeling images taken in an ESEM. The very slow peel rate of 1 μm/s ensured the failure mode was interfacial. Two major features are apparent. First, the peel front is very ragged. The PSA seems much more strongly attached to some locations than to others. The strongest adhesion led to the formation of long PSA strands (called fibrils in the PSA industry [20]). Also evident are cavities in the PSA layer formed by a process called cavitation. Viewing the complexity in these images or in the parent video clip emphasizes that continuum models based on isotropic materials are unlikely to give realistic descriptions of PSA peel behavior from paper.

FIGURE 7. A TEM photograph of tape 411 after peeling from Copy Paper 1. Individual fibers picked from the paper are visible and one has produced a neck of adhesive during peel.

DISCUSSION

Tape peeling experiments from paper are attractive because: they require simple equipment; they give reproducible results; and, they reveal something about the properties of the PSA, the paper and, in some cases, both. However, detailed analysis is difficult. Energy is consumed by bending the tape backing and paper, by deforming the PSA and by propagating the crack front. Indeed, much of the peel work is consumed by viscoelastic flow. The existence of flow is emphasized in Figure 9 which shows PSA fibril formation at the peel front. Although we can precisely measure the overall peel force, it is difficult to obtain the corresponding force applied at the paper surface by the PSA. However, the force experienced by the paper surface can be varied. For a given PSA in peel, increasing the peel rate or decreasing the temperature will increase the force applied to the paper/PSA interface.

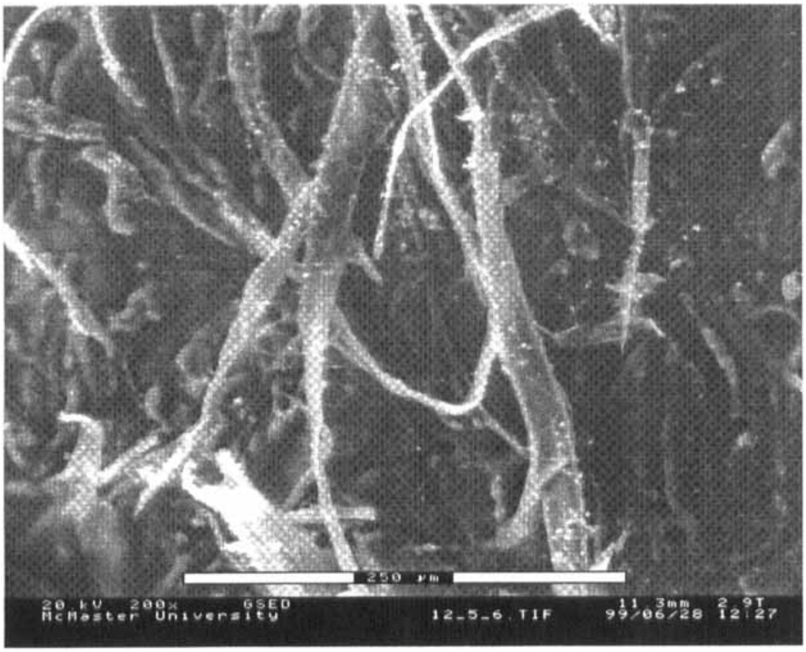

FIGURE 8. A SEM picture of a tape 411 after peeling from Copy Paper 1. The area to the left shows a layer of fibers embedded in the PSA, whereas the right hand portion is fiber free. The indentations in the PSA layer caused by fibers before peeling are visible on the right.

The most common failure modes in our work were (pseudo) interfacial and paper failure (delamination). In the case of paper failure, it seems clear from the literature that the steady-state delamination force gives a measure of the z-direction paper strength [21, 22]. In the case of interfacial failure, the peel force reflects mainly the PSA rheology. As mentioned previously, the highest sustained peel forces were for interfacial failures, whereas the paper delamination required a relatively small force. To maximize peel force, paper delamination must be avoided.

Perhaps the most fascinating aspect of tape peeling from paper is the transition from interfacial to paper failure. This transition was observed when the peeling stresses at the PSA/paper interface were increased by: 1) increasing the peel rate, which, for a viscoelastic PSA, generates increased force at the interface; 2) increasing the lamination force, which, in turn, increases the PSA/paper contact area and, thus, the ability of the PSA to transmit stress to the fiber network; and 3)

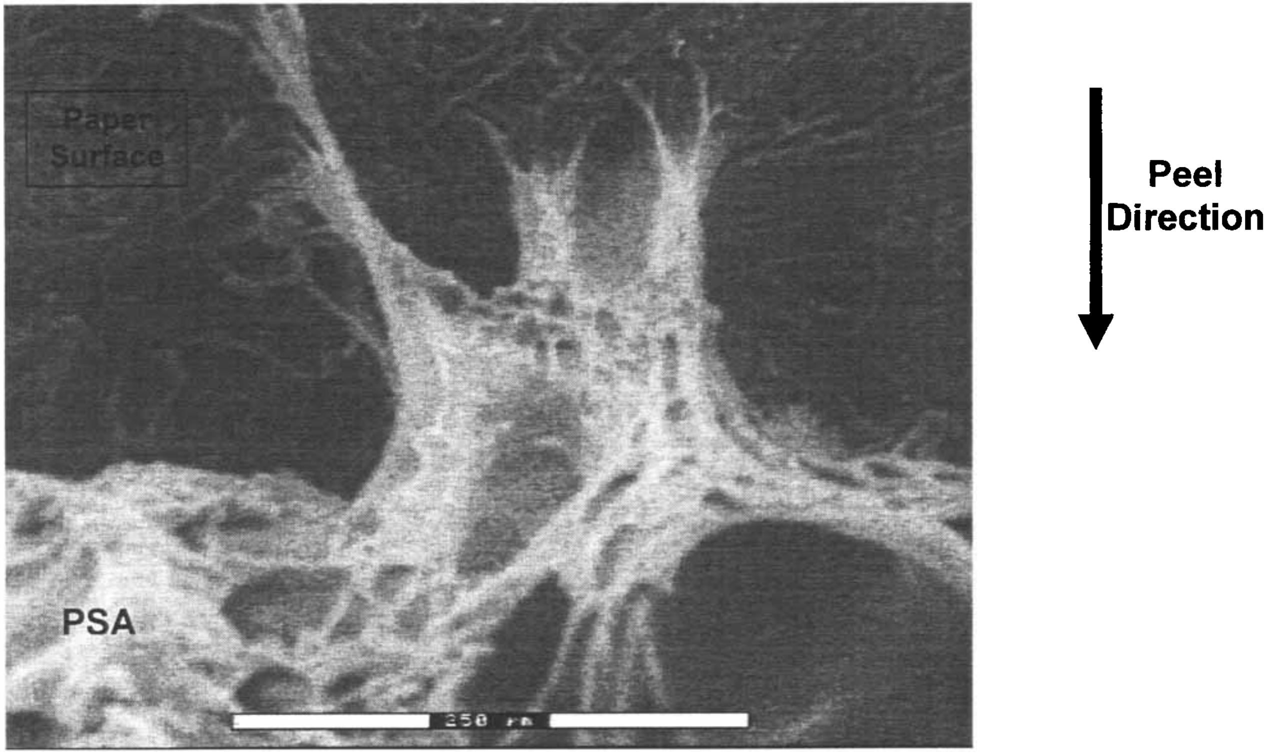

FIGURE 9 A video frame from peeling (180 deg) 3M 906 tape from copy paper in an ESEM. The peel rate was 1 µm/s and the horizontal bar corresponds to 250 µm. The PSA elongates to form fibrils and cavitation generates bubbles in the PSA during peeling.

changing the PSA/cellulose interfacial energy by surface modification. From these studies we can formulate some generalizations.

A) Paper roughness/porosity or sheet density seems to be the best predictor of the propensity towards paper failure.
B) Hydrophobic treatment of handsheets did not lower the tendency for delamination. The role of paper surface energy may be more important with smooth papers. Indeed, release papers are extremely smooth in addition to being treated with hydrophobic agents. Usually the surface smoothness is achieved with a coating.
C) Increasing lamination pressure increases the peel force required to give interfacial failure. However, the probability of paper failure increases with the stress applied to the paper surface and, thus, with the lamination pressure.

The onset of paper delamination is a fracture process. In cases of mixed paper failure the peel starts at a single location in the paper surface (see Figure 2). Thus, delamination starts at an initiation site, which we consider to be a flaw. Although we have yet to observe the onset of delamination directly at a microscopic level, it seems reasonable to speculate that the flaw is associated with a fiber end – since we routinely observe fines and fillers in the peeled adhesive layer after interfacial peeling, we presume that fines or fillers cannot convey enough stress to the fiber network to initiate delamination. Furthermore, we can speculate that the most active fiber ends with respect to delamination have the following properties. 1) The fibers are oriented in the peel direction with the only active end (with respect to crack initiation) facing the oncoming peel front; and, 2) the fiber end presents a significant surface area for interaction with the PSA. In other words, a high local roughness near the fiber end should give the highest chance for initiation of delamination. Hiding the fiber ends with a coating should prevent catastrophic delamination; however the coating may be removed in peel.

When considering how a specific PSA tape interacts with a variety of papers, a useful parameter is the critical peel rate, r, which we define as the minimum peel rate required to induce paper delamination. Of course r will depend upon the experimental variables including the tape/paper lamination pressure, lamination time, dwell time (*i.e.*, age of the bond) and peel angle—for discussion purposes these variables are assumed to be constant. The greater the r value, the greater the resistance of the paper to delamination.

We propose that two independent sets of paper properties (*i.e.*, principal components) are required to explain/predict the response to a specific tape in peel. The first component is the surface energy of the paper. The higher the surface energy, the higher will be the corresponding work of adhesion which has been shown to correlate with practical adhesion in the case of toners [18]. PSA/paper adhesion is required to transmit peeling stress to the paper. Although pure cellulose has a relatively high surface energy, pitch from wood and added sizing agents can give substantially lower surface energies [15, 23].

The proposed second component required to explain the response of paper to peeling is paper structure. This term includes the paper surface's cohesive strength and those factors influencing the total PSA contact area with paper. Yamauchi et al. proposed that the tendency of paper to delaminate in peel correlated with paper density [6]. For a given type of paper, strength increases, whereas porosity and surface roughness tend to decrease, with increasing paper density. Figure 10 shows qualitatively how this two-component analysis can be applied to our results. Filter paper has a low density and a high surface energy. Thus, filter paper sits in a corner of the three-dimensional space and the r values for filter paper are low. Glassine is smooth with a high density and has a high surface energy. Thus, glassine sits in another corner which has a high r value. Release papers, such as tape backings, are smooth, dense and often have a silicone surface coating to give low surface energy. In this corner the r value is infinite since paper delamination should never occur. The fourth corner (upper right) corresponds to low density, low surface energy papers which are not common. We prepared such a material by the fluorochemical treatment of handsheets – they have an intermediate r value. Copy papers, where the transition from interfacial to paper failure is easily observable, are in the center of this space. Their surface roughness and surface energies are both intermediate.

Our analysis shown in Figure 10 is qualitative. However, a PSA manufacturer wishing to characterize a tape product could do this quantitatively. Principal component statistical analysis could be applied to data for a specific tape to relate commonly measured paper properties to these axes. In such an analysis, surface roughness, Scott Bond energy, density, and PSA lamination pressure are all likely to contribute to the paper structure term.

In ongoing work we are developing a mechanistic model to predict the tendency for paper delamination in peel. When completed, this analysis will illustrate the role of fiber length, fines, roughness and peel mechanics.

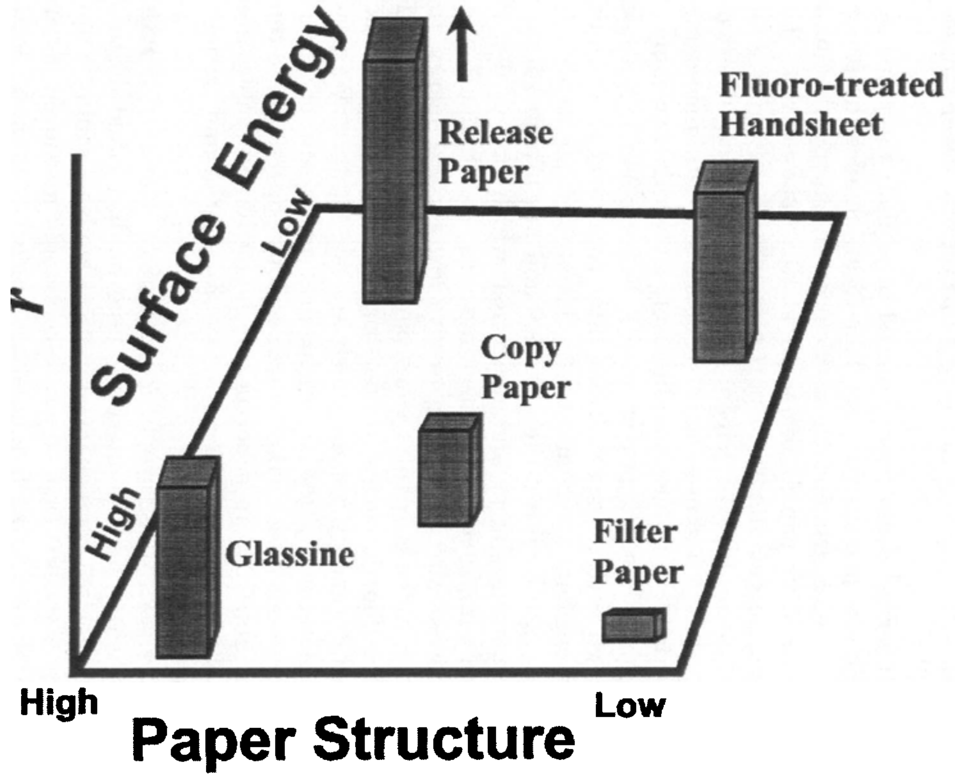

FIGURE 10 Illustration of the use of paper structure and surface energy to categorize the tendency of paper to delaminate in peel. The z-axis shows ***r***, the minimum peel rate required to induce paper delamination – the greater the value, the stronger the paper is in respect to this failure mode.

CONCLUSIONS

1. For most types of machine-made paper it is possible to observe both interfacial peel and paper failure depending upon the type of tape, the lamination pressure, the peel rate and the peel angle.
2. The tendency towards paper failure increases with peel rate and tape/paper lamination pressure.
3. For a given paper and tape, the maximum peel force is observed with interfacial failure. Paper delamination forces are relatively low. Thus, if a high peel force is desired, paper delamination should be avoided.
4. Surface roughness and paper density are predictors of whether paper failure will occur. In the extreme, our glassine samples have never given paper failure in this work, whereas poorly beaten handsheets or filter paper nearly always give paper failure on peel.
5. Machine-made paper is exceptionally anisotropic in its response to peel. The peel conditions for onset of paper failure depend on peel direction. Machine direction *versus* cross direction peeling gives different results. Furthermore, when peeling in the machine direction, "towards the dry-end" *versus* "towards the wet-end" peeling also gives different results.
6. The onset of paper failure is remarkably insensitive to surface energy. Contrary to intuition, we saw little evidence of lower peel forces or a lower tendency towards paper failure when paper was given hydrophobic treatments. On the other hand, hydrophobic treatments do lower the steady state peel force during interfacial failure.
7. We propose that long fibers are responsible for the initiation of paper failure. Fines and filler can give weak peel due to weak boundary layer failure. However, small particles are not sufficiently connected to the fiber network to initiate delamination.

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