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Fracture of Xerographic Toner on a Swelling Substrate

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## FRACTURE OF XEROGRAPHIC TONER ON A SWELLING SUBSTRATE

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### ABSTRACT

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Repulping of toner-printed paper produces a slurry of toner particles and fiber from which the toner is then removed. It has been shown that the ability to remove the toner is affected by the size and shape of toner particles. The subject of this work is the effect of the mechanical properties and thickness of toner layers on the breakup of the toner due to paper swelling.

Uniform crack spacing is shown to occur in toner lines printed on both paper and cellulose acetate films upon immersion in water. Presented here is a model to predict the crack spacing of a toner line from the thickness of the line and the relative cohesive and adhesive strengths. Experimental results show the crack spacing to increase linearly with thickness as predicted by theory.

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### INTRODUCTION

Repulping of toner-printed paper produces a fiber slurry with plate-like toner particles (one dimension much smaller than the other two). These particles have been difficult to separate from the fibers in conventional deinking processes because of their shape, a wide size distribution (longest dimensions), and fibers remaining attached to the particles [1-9]. It appears that the best way to control the particle size and shape is to either agglomerate the particles to a larger, three-dimensional shape [10] or to use excessive mechanical treatment to break the particles down to a smaller size [11]. A better understanding of how the toner fractures into particles should lead to more efficient processes for producing particles of an optimum size.

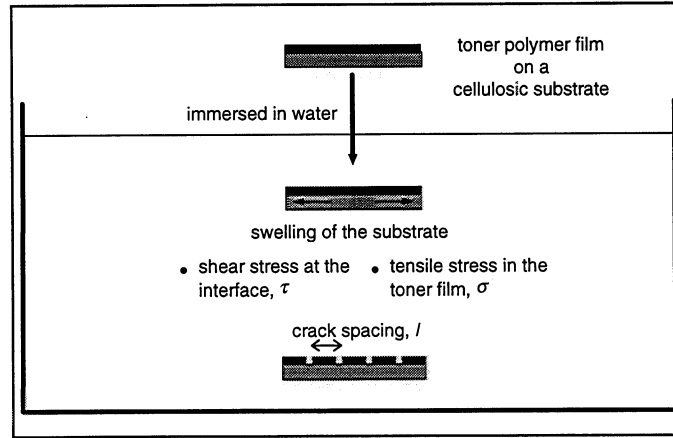
Vander Wielen *et al.* showed that significant fracture of toner may occur upon immersion of toner-printed paper in water, without any external mechanical forces [12]. Fracture occurred as a result of the mechanical stress between the swelling paper and the nonswelling toner. It was observed that fracture in a printed line of toner was characterized by regular spaced cracks. The crack spacing should give an approximate measurement of the toner particle size produced through a swelling mechanism.

It will be shown in this paper that by considering the toner on paper to be a uniform two-layer composite, the regularly spaced fracture of a toner line may be expressed in terms of mechanical properties and a critical crack spacing. Measurements of the crack spacing of a line of toner from a model system (toner on cellulose acetate) will be compared with actual toner-printed paper.

### FRACTURE OF A BRITTLE FILM ON A SWELLING SUBSTRATE

A composite of a nonswelling film (representing toner) bonded to a swelling substrate (representing paper) is shown in **Figure 1**. A significant difference in swelling between toner and paper is expected based on the significant difference in moisture sorption (typical toner polymers absorb < 1% water [13]). The difference in swelling produces shear stresses at the interface that transfer tensile stresses to the film [14]. The shear stress is greatest at the ends of the film, then decreases, while the tensile stress in the film is lowest at the ends and increases along the length. These stresses can lead to fracture of the film with a regular crack spacing,  $l$  (**Figure 1**). Fracture with a regular crack spacing has been previously observed in brittle films on strained substrates that are ductile and have a much greater thickness than the film [15-19]. This fracture is characterized by sharp, uniformly spaced cracks that are perpendicular to the applied strain.

This fracture behavior can be explained by the Kelly-Tyson model [14]. This model was originally developed for fiber-reinforced composites, but it has been applied to thin films [15-19]. This model assumes that the interface is plastic and the interfacial shear stress equals the yield strength of the interface,  $\tau^*$ . The shear stress is constant over a length,  $L/2$ , over which the tensile stress increases linearly until it reaches the tensile strength of the material,  $\sigma^*$

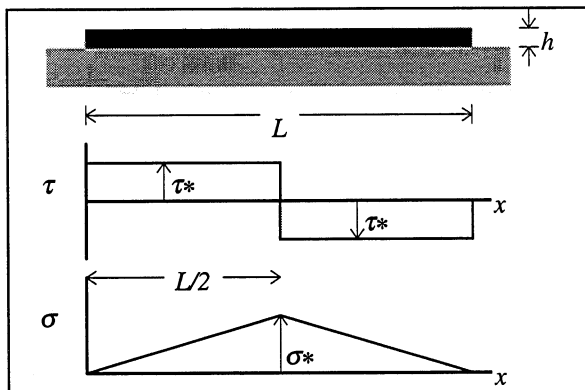


**FIGURE 1:** Swelling of the substrate can fracture the brittle, nonswelling film into multiple segments. The fracture can be quantified by a crack spacing,  $l$ .

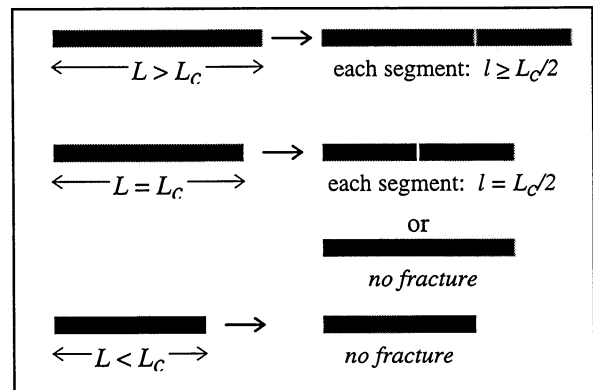
(**Figure 2**). A section of film will fracture into shorter segments as long as the tensile stress within each section reaches the strength. If the section is not sufficiently long for the tensile stress to reach the strength, fracture no longer occurs. The minimum length for the tensile stress to reach the fracture strength is  $L_c/2$ ; thus, the minimum section length that will fracture is  $L_c$ , the critical crack length. A linear relationship between the critical crack length and thickness is predicted from the model, proportional to the tensile and interfacial shear strengths:

$$L_c = 2h \frac{\sigma^*}{\tau^*} \quad (1)$$

where  $L_c$  is the critical crack spacing [mm];  $h$  is the thickness of the toner layer [mm];  $\sigma^*$  is the tensile strength of the toner film [MPa]; and  $\tau^*$  is the interfacial shear strength [MPa]. The actual crack spacing,  $l$ , should be distributed between  $L_c/2$  and  $L_c$  (**Figure 3**) [15, 16]. Segments with a length greater than  $L_c$  are likely to fracture, while segments with a length less than  $L_c$  are not. There should be no segments with a length less than  $L_c/2$ , as this is the minimum length for the tensile stress to reach the fracture strength (see **Figure 2**).



**FIGURE 2:** Distribution of interfacial shear stress ( $\tau$ ) and film tensile stress ( $\sigma$ ) along length of film on a swelling substrate. Both stresses will reach a maximum value corresponding to the interfacial yield strength,  $\tau^*$ , and the tensile fracture strength,  $\sigma^*$ .



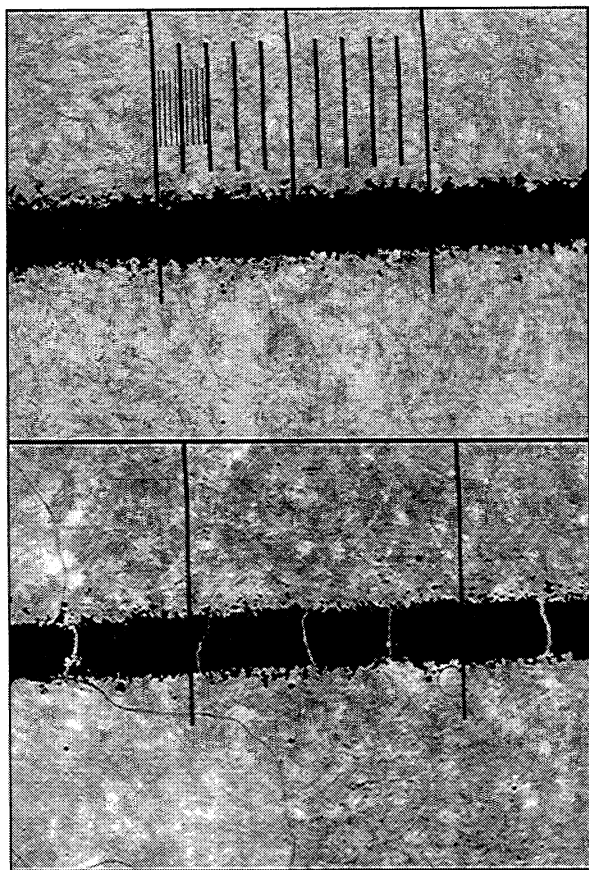
**FIGURE 3:** Sections of film (left) will fracture into smaller segments depending on the length of the section,  $L$ . The film will fracture until the crack spacing,  $l$ , is between  $L_c/2$  and  $L_c$ .

## EXPERIMENTAL

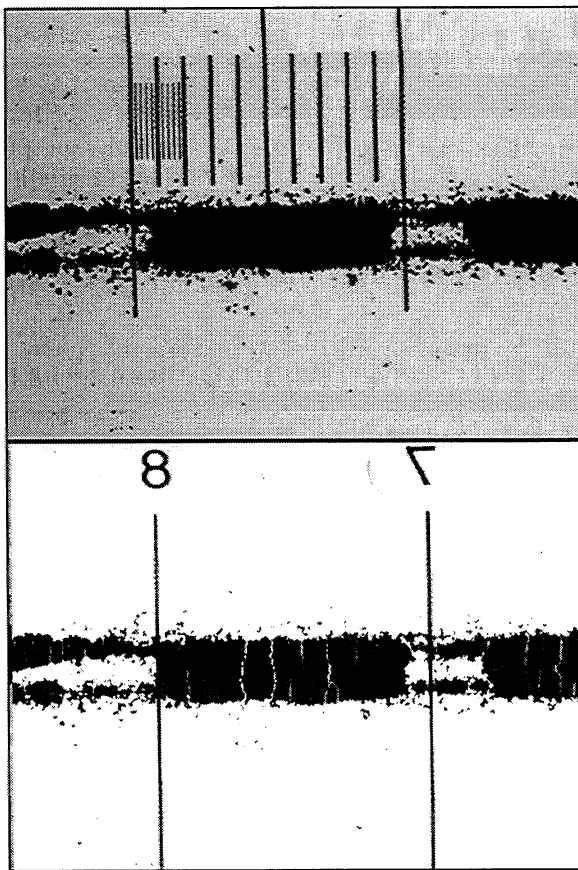
Toner lines (0.5 point) were printed on both paper and cellulose acetate from a laser printer (toner composition: 40-50% styrene acrylate resin, 45-55% magnetite, 1-3% asine dye, 1-5% polypropylene, 1-2% silica). Both the paper and cellulose acetate substrates were printed by taping a section (12.5 mm x 30 mm) of the substrate to a sheet of paper, and then running this assembly through the printer. Multiple layers of a line were produced by repeating the printing on the same sample. These lines were all printed in the cross direction of the paper, because Vander Wielen *et al.* showed that a line of toner preferentially fractured perpendicular to the cross direction [12]. The thickness of the printed toner line was determined by subtracting the thickness of the unprinted substrates from the total thickness of the substrate and printed line. The thickness was measured with a flat face electronic micrometer (Mitutoyo 293 Series, resolution +/- 0.001 mm, accuracy +/- 0.002 mm).

The paper was standard 20 lb xerographic paper stored at 23°C and 50% relative humidity (103 ± 1 micron thick). The film of cellulose acetate (Aldrich, average  $M_n$  ca. 30,000 (GPC), 39.8 wt% acetyl content) was produced in our laboratory by casting on a Teflon surface from an acetone solution. The technique produced flat films (124 ± 2 micron thick) that remained flat when immersed in water, indicating that they contained no significant residual stresses. The cellulose acetate films were dried to constant weight in an oven and stored in a desiccator until just prior to use.

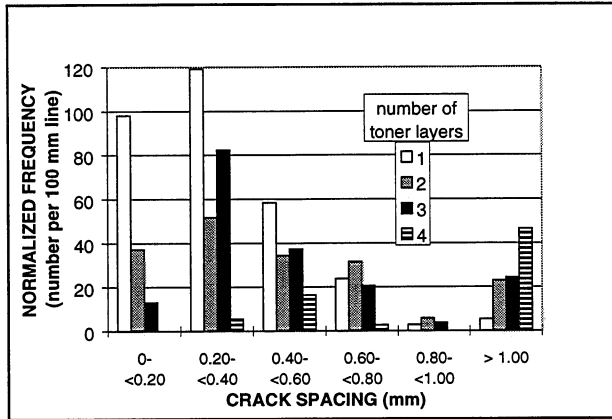
The substrates that were used originated from the same sheet of paper or same film of cellulose acetate. For each value of average crack spacing at constant toner thickness on **Figure 9**, three printed substrates (approximately 20 mm of printed line each) were analyzed. The average crack spacing is the average from all the measured lengths of crack spacing at the constant toner thickness. For example, each data point in **Figure 9** represents the average of 25 to 350 cracks measured on the different printed substrates.



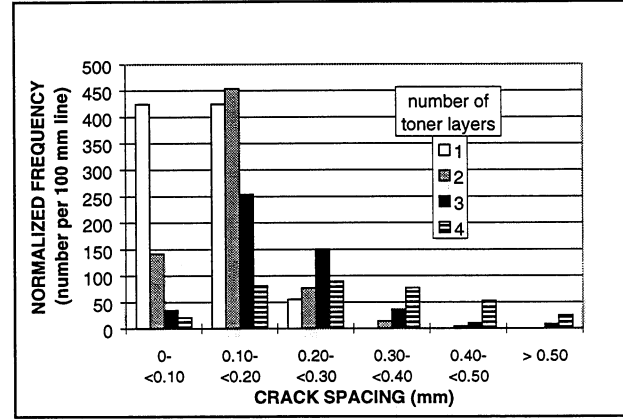
**FIGURE 4:** Optical micrographs of single-layer toner lines (0.5 point) printed on paper before (top) and after (bottom) immersion in water. The scale is 1 mm.



**FIGURE 5:** Optical micrographs of single-layer toner lines (0.5 point) printed on cellulose acetate before (top) and after (bottom) immersion in water. The scale is 1 mm.



**FIGURE 6:** Distribution of crack spacing in a 0.5-point toner line printed on paper from a laser printer. The normalized frequency represents the number of particles in each size bin for a 100-mm line.



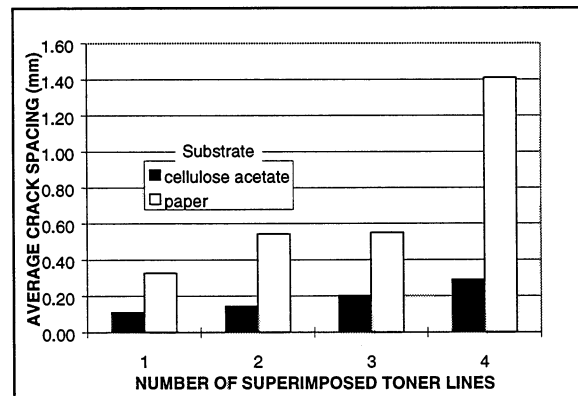
**FIGURE 7:** Distribution of crack spacing in a toner line printed on cellulose acetate. Note the change in the scale of both axes from Fig. 6.

The printed substrates were immersed in deionized water at 25°C. The spacing of the cracks that formed was measured from acquired digital images using image analysis software (OPTIMAS). Crack spacing was measured as the distance between cracks. The frames were calibrated with an optical micrometer. The photographs of the magnified samples were taken with a 35-mm camera (OLYMPUS C-35A) mounted on a microscope (OLYMPUS BH-2). The digital images for image analysis were acquired with a microscope (Zeiss Axioskop) and a digital camera (Sony 3 CCD). Transmitted light was used for both. The immersed samples were placed between glass slides during observation to keep the sample flat and wet.

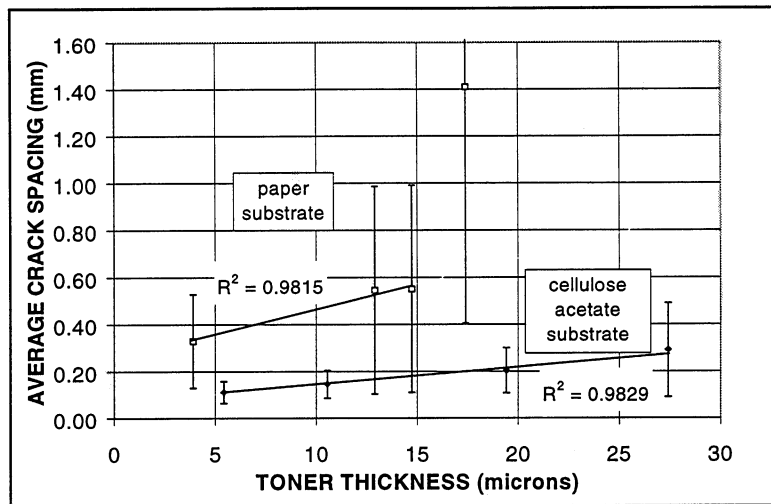
## RESULTS AND DISCUSSION

Figures 4 and 5 show that the single-layer lines printed on paper and cellulose acetate are similar before and after immersion in water. The lines have a consistent width and a continuous appearance. After immersion, parallel cracks form across the width of the line and the segments of line remained attached to the substrate. Figure 5 shows that the lines printed on cellulose acetate did have sections that were not continuous across the width. Only the sections that were continuous were used for crack spacing measurements. From these figures, it can be concluded that the model system (toner on cellulose acetate) approximately reproduces the physical structure and fracture behavior of toner on paper.

Figures 6 and 7 show the crack spacing distribution for printing multiple times on both substrates (increased toner thickness). Printing additional lines shifts the spacing distribution to larger values. Additional printing increased the measured thickness of the toner line by superimposing lines on one another. Lines that were printed multiple times



**FIGURE 8:** Average crack spacing for multiple toner lines on both paper and cellulose acetate.



**FIGURE 9:** Average crack spacing for the measured toner thickness on both paper and cellulose acetate substrates. The error bars are 1 standard deviation of the spacing measurements.

were checked carefully using the optical microscope to make sure the lines were superimposed. A thicker line is expected to have a greater crack spacing (Eqn. 1). **Figure 8** shows the increase in the average crack spacing.

**Figure 9** shows that there is an excellent linear fit between the average crack spacing and toner line thickness for both substrates, except for the thickest (4 layer) toner line on paper. A linear relationship is predicted from the Kelly-Tyson model, where the crack spacing is proportional to the ratio of strengths,  $\sigma^*/\tau^*$ . This indicates that the crack spacing in a toner line can be determined if values of  $\sigma^*$  and  $\tau^*$  are known. The critical crack spacing may approximate the maximum size of toner particles that remain in this system after substrate swelling. A thicker ( $h$ ) or stronger ( $\sigma^*$ ) toner line or a line with a weaker interface ( $\tau^*$ ) will promote larger particles.

One of the key assumptions for applying the Kelly-Tyson model is that the interface deforms plastically. An interface between materials can be considered to consist of three regions: the actual microscopic interface and each of the immediately adjacent materials [20]. Therefore, both the cellulose acetate and paper can be considered part of the interface, and it is reasonable to believe that either can yield plastically under these high moisture conditions. In addition, observations of the samples showed that there was no clean delamination of the toner from either substrate. The particles were hairy, as has been previously described [4-6]. Clean delamination would indicate a brittle, adhesive fracture rather than yield.

The crack spacing for the thickest (17 micron) toner line on paper increased abruptly from the previous points, deviating significantly from the predicted linearity (**Figure 9**). It is likely that the crack spacing exceeded an upper limit for the model. The toner line became too thick for the tensile stress to reach the fracture strength at a regular spacing as predicted by the model (see **Figures 2 and 3**). It is possible that the length over which the interfacial shear stress can effectively transfer tensile stress to the film was exceeded. Therefore, the tensile stress does not increase as much as expected. Fracture still did occur but with a wider spacing and a much greater variability. The samples printed on cellulose acetate remained linear and followed the model at a greater thickness because the upper limit for crack spacing had not been reached.

The toner lines on paper showed a greater crack spacing than the lines on cellulose acetate. The greater spacing may be due to the lower strength of the toner/paper interface on a macroscopic level. From **Figure 9**, the strength ratio (slope),  $\sigma^*/\tau^*$ , is greater for paper. Assuming the toner strength,  $\sigma^*$ , to be the same in both systems, this indicates a lower interfacial shear strength,  $\tau^*$ , for the toner/paper interface than the toner/cellulose acetate interface. It is reasonable to believe that the toner/paper interface should have more weak points for failure because it is more discontinuous. Cellulose acetate is a continuous material and has an essentially planar interface with the toner. Paper is a fibrous network with an irregular surface and irregular interface with the toner. Therefore, shear stress can be concentrated in smaller areas of contact, and failure can occur at a lower overall shear stress.

## CONCLUSIONS

- A printed toner line on a swelling substrate fractures with multiple parallel cracks, which can be quantified by crack spacing. This process limits the maximum particle size that can be obtained from deinking.
- The crack spacing increases linearly with thickness for our model system (toner/cellulose acetate), as predicted from the Kelly-Tyson model. For toner-printed paper, the same relationship holds within practical toner thickness limits. The crack spacing of toner on paper deviates from linear behavior at the largest toner thickness because the tensile stress in the thicker line does not reach the fracture strength at a regular spacing as predicted by the model.

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