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On the in-plane distribution of filler in paper

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Formation, Filler Distribution, Statistics, Retention Mechanisms.

Abstract/Conclusions

The in-plane distribution of filler in paper should be of high interest for a practical reason. High variations in filler loading could cause weak regions at peak loads, as well as low opacity regions at the "valleys" – improvements in the uniformity of filler loading could lead to an improved optimum balance between strength and optical properties. This would in practice allow higher average fiber replacement by filler.

Despite such significance, few studies of the in-plane "filler formation" have been published. We will briefly discuss those prior publications available to us, the methodology for and some difficulties with the physical measurement, and examine commercial copy paper as well as handsheets made with pulsating drainage at headbox consistency.

The distribution of filler and fiber in these samples is analyzed in various ways and the analyses are compared. A quick and simple analysis based on cumulative density distributions gives an attractive but apparently false result, due to a high level of stochastic noise. A more elaborate analysis of correlating two images by windowing is reported, and shows a linear fiber-filler relationship with no long-range influences in the plane of the paper. The correlation found is not very good, but an application of neural network analysis shows that the addition of nonlinearity to the modeling will not improve the results to any significant degree – this is a case where classical linear analysis is quite sufficient.

The correlation improves as first pass retention is increased by use of additives – prebonding filler to fiber strongly is clearly detected in the statistics in a way that is consistent with this retention mechanism.

While these studies provide some fundamental information related to retention mechanisms and sheet structure, the methodology presented provides distributions and relationships necessary for estimating what could be gained by reducing the levels of variation, and may in the future contribute to maximizing fiber replacement by filler.

Introduction

Little prior work seems to be available on the in-plane distribution of filler – reported filler distribution determinations mostly deal with the thickness or z-direction. Corte [1] removed by chemical dissolution chalk filler from a highly filled handsheet (40% chalk) and found a negative correlation of chalk mass to fiber mass, indicating that excess chalk filled in low fiber density areas making the total mass distribution more uniform. The measurement was based on beta radiation with 1 mm geometric resolution. The found negative correlation is not explained. Norman et al. [2] use a similar method with paper from a pilot machine. While Corte analyzed the fiber and filler distributions by calculating correlations of pointwise observations, Norman et al. resorted to an inspection of power-spectra calculated from two arrays of numbers (discrete images of total mass and fiber mass). Visually the two spectra appeared to be of rather similar shape, which was interpreted as an indication that the local filler mass is proportional to the local fiber mass. Sampson and Turner [3] report on handsheets formed at fiber concentrations ranging from 0.017% to 0.12% in a standard British sheet former. The GCC filler was again dissolved, while the imaging was done by optical means. They found that the ratio of variances before and after filler dissolution was quite different for softwood and hardwood pulp – so also was the ash content of the sheets. They suggest concluding that small variance (good sheet uniformity) will cause a high retention level. Davidson [4] and Bown [5] have reported macroscopic observations, not based on any sort of imaging, using filler dissolution techniques. These references discuss the interaction of fillers with fiber material, and the consequences on optical and mechanical properties of paper.

Only Corte reported conventional statistical analysis, in the form of correlation between pixelwise matched images (i.e., matching individual elements of the two observed arrays). A two-dimensional window variant of moving average smoothing before computing the correlation apparently gave results quite similar to the pointwise calculation. A precondition for pointwise or windowed analysis of the relationship between fiber mass and filler (or total) mass is that the sample is well enough aligned and undeformed after filler dissolution for remeasurement of the same exact sample points. This alignment problem is avoided by comparing the distributions observed before and after filler dissolution, as was done in references [2] and [3]. Our analysis below suggests that a comparison of distributions can give misleading results, and even with carefully aligned samples, statistics must be pursued far enough to get correct results.

Theoretical

The question posed is "how does the total mass at a pixel depend on the fiber mass in some neighborhood of this pixel". We increase the model complexity until there are no further gains in goodness of fit. Analysis of cumulative distributions. A monotonically increasing relationship between two random variables can be solved, if the cumulative distributions are known. If $y=f(x)$, then the solution is $f= F_Y^{-1} \circ F_X$ where the superscript "-1" indicates inverse mapping, not exponent. The hidden assumption is that the relationship is not noisy but one-to-one: once either variable is randomly sampled, the other is completely determined. Noise in the model $y=f(x)+e$ will not allow solving f from the distributions without further *a priori* knowledge. Pixelwise analysis. The noisy model yields to regression analysis, which can be carried out in a spreadsheet by rectifying the measured matrices each to a column. Multilinear regression. Pixelwise analysis assumes no interactions from neighboring areas on the ash retention at a given pixel. To account for short-range influences, the total mass at a given pixel is modeled as dependent on a "window" of fiber mass pixels around it. Pixels on or close to the border of the measured area are discarded from the analysis. The sum of coefficients in the multilinear model shows the effect of an increase in average fiber mass on total mass. Nonlinear modeling. A neural network model extends on the multilinear ones, by allowing nonlinearity. This enables checking whether nonlinearity will significantly affect the goodness of fit.

Experimental

Commercial copy paper and four types of handsheets made with the pulsating handsheet forming device MBDT were studied. An earlier version of the MBDT and its application are described in [6,7, and 8]. The mass imaging was done with IPST's beta radiation based formation tester, with 1 mm geometric resolution, measuring a 50 mm x 50 mm area to give 2500 pixels. Valley refined hardwood and softwood pulps were mixed in 60/40 ratio for the handsheets. The retention of PCC (ash content of final handsheet) was adjusted with additive dosage, with otherwise unchanged furnish. The PCC was dissolved with HCl from paper samples compressed between 150 mesh screens to prevent deformation during dissolution, rinsing or drying. The dried samples were carefully realigned for measurement of the same physical pixels: the alignment was better than 0.5 mm, based on cross-correlations between rows and columns before and after dissolution.

Results and Discussion

Denoting the total mass by y and the fiber mass by x, our linear models are of the form y=a.x+b where a and x can be vectors. Denoting the sum of components of a by A, the numerical values found for copy paper were A=1.15 from cumulative distributions, A=0.80 from linear regression, and A=0.998 from multilinear regression with 3x3 window (very similar result also in terms of R^2=0.67 was obtained with a 5 x 5 window). Neural net modeling with a 5 x 5 window gave a slight improvement of R^2 to 0.7 only, indicating that nonlinearities can be neglected, while pixelwise regression gave a poor R^2=0.51 showing that short-range effects need to be observed by windowing.

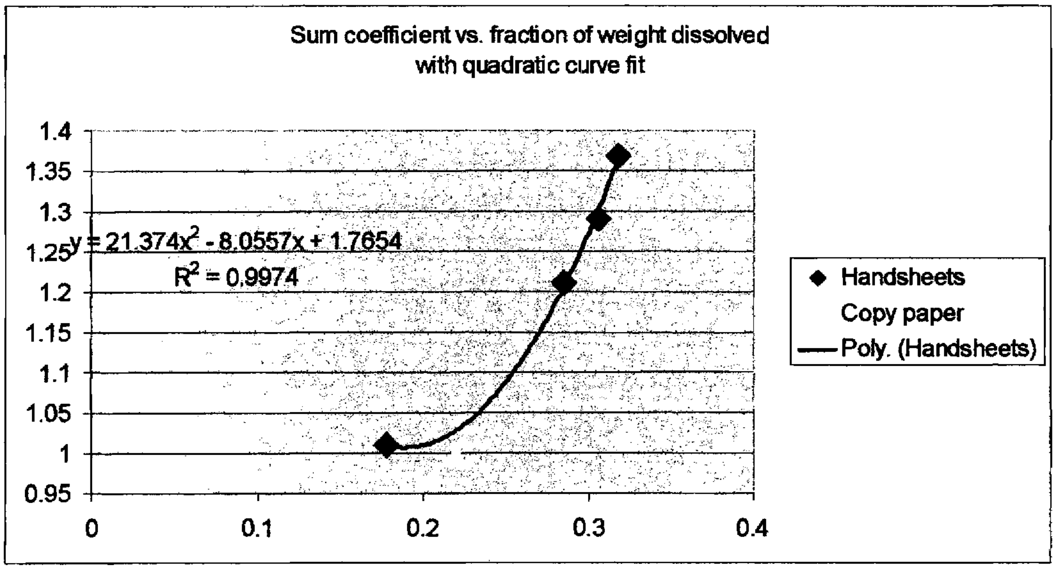

Figure 1. As retention chemical dosage is increased forcing FPR towards 100%, the PCC attached to fibers amplifies the in-plane variations of fiber mass. This amplification factor is shown against the fraction of mass lost during acid dissolution of PCC and the following washes and rinses with water.

Now we have good grounds to state that multilinear modeling is the most correct approach, and its A=0.998 show that the total mass locally matches the variations in fiber mass. This means that the PCC filler neither plugs holes in the fiber matrix, nor follows the fibers – it appears to find its in-plane distribution independently of the fiber mass in the commercial copy paper. Note that the simpler statistical analyses would suggest the filler "going where there is more fiber" with A=1.15, or "plugging holes" with A=0.80. From the same data, we could falsely find a dominating retention mechanism if we did not diligently extend the analysis until no further gains in model quality (goodness of fit) are found. The multilinear analysis was also carried out with measurements from handsheets, where the PCC filler content was varied by changes in retention chemistry. As expected, a high level of retention is achieved by fixing the PCC to the fibers (or fibrils) prior to forming. This leads to a correlation of the in-plane distribution of filler with fiber, as shown by the sum coefficient A. The handsheet measurements seem to match well the results from commercial copy paper. The weight fraction lost by acid dissolution of PCC and washing matches the separately measured PCC content (by titration) to within 1 percentage point; also other materials are lost but their contribution is not significant for our handsheets. However, the about 22% fraction lost with commercial copy paper is clearly higher than its ash content, and the corresponding data point could be shifted left in Figure 1. This would only bring it closer to the handsheet observations.

Careful sample alignment and multilinear regression analysis combined with the filler dissolution technique appear necessary and sufficient for proper analysis of the mechanics of retention, while an ad hoc statistical approach will possibly give misleading results.

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