# Rheomembrances

## Börje Steenberg

Paper Technology, Dept. of Fibre and Polymer Technology, Royal Institute of Technology, S-100 44 Stockholm, Sweden

## ABSTRACT

A personal report of the early rheology studies of papermaking and their technological impact.

## INTRODUCTION

Specification of paper was in the old times related only to its basis weight and a general similarity to a given paper sample. Paper makers had agreed never to guarantee any other quantitative figure, The reason was simple. They could not control their production due to the almost complete absence of measuring instruments on the Any laboratory test figures machines. arrived only when a new order was on the machine. Further paper was then hardly ever used for purposes where strength properties were critical. Flexible thin paper was used for writing and printing and sometimes to twist a cone. Board qualities were used for hard cover of books and some building use, like roofing paper impregnated with asphalt and tar.

It was only with the increasing use of mechanical pulp (groundwood) that quantitative testing of paper became important. The German State authorities were the first to react to paper products with groundwood, which had such low strength and even lower durability that they could not be used for archival documents.

In 1884 the first "Abteilung für Papierprüfung" was started at the German

"Mechanisch-Technischen Versuchsanstalt" in Charlottenburg. The institute, later known as the "Reichs Physikalische Prüfungsanstalt" in Berlin—Dahlem had the purpose to lay down rules for paper qualities to be legally prescribed for various public documents. Such "normal paper" was introduced in Sweden in 1907.

For standard types of writing and printing papers properties like smoothness and opacity was important. Typically folding strength was given priority to tensile tests. The suggested mechanical testing methods were named after their inventors. The most common test was the Mullen test, a burst test. The testing machine, hand driven, was simple, but the theory behind is still not worked out. Walter Brecht, the famous paper professor in Darmstadt has told me than when he was first hired as teacher in paper testing there were at least 50 tests for stiffness and the same number for water resistance. Ink was then a watery solution. The paper testing methods were essentially unchanched from the first monograph in the filed (Papierprüfung by W. Herzberg 1920) until after the second war.

Newly developed strong kraft papers and board around 1925 suddenly increased the field of use of paper. Paper bags and sacks and stiff boxes of paperboard became common. Mechanical testing was now much more important and new paper testing methods were suggested. Some large international paper merchants built their own paper-testing laboratories to control their supplier's product. A leading example of this was Buhrmann in Amsterdam and the Danish Newsprint Buyers Association's laboratories in Copenhagen. Both developed new tests, some still used today.

When I was hired to lead the new Paper Technology Department at the Swedish Forest Products Laboratory "STFI" in 1943 paper testing had reached an importance it never had before. The reason was the war and the lack of textile fibres. All Europe harvested with machines producing sheafs, but the necessary binding strings, normally of sisal fibres, was un-obtainable. The solution was strings of twisted paper.

Swedish kraft Yankee machines got orders for such paper for export. The trouble was that many boats were torpedoed before the quality of the paper could be tested. So the tests were made at the Royal Institute of Technology in Stockholm at the "CCL" laboratory. If acceptable the paper was paid before it left a Swedish harbour.

Our first work at STFI was obvious: improving the wet strength of twisting paper by adding polymers, in the first instance melamine<sup>1</sup>.

The twisting string paper required not only a very high machine direction "MD" tensile strength but also a good breaking elongation in cross direction "CD", which was difficult to measure with the existing testing machines. Paper elongates at break very little and its importance was neglected.

The trick to make the paper was to" beat" the unbleached sulphate pulp and do the milling at low temperature at slightly elevated pH. The wire part of the PM was then run at higher speed than the jet from the headbox. This was known to produce high tensile strength. in the MD direction. Possibly more than 30 PM in Sweden produced this paper grade during the war, mostly at wire speeds under 150 m/min.

STFI, at this time a government research institute, was run as a university. Being new to the industry I visited as many mills as possible and spoke to the beater foremen and the machine tenders to find out the secrets of the trade and to formulate my research program. I found that the properties of the twisting paper from different factories was different, possibly due to varying paper machine designs. This was against the common thinking, according to which the paper was "made in the Hollander", the machine in which the pulp was milled. Paper properties were thought to be in the hands of the beater foreman. It took me at least 30 years to understand the physics of the beaterman's grab test, then a critical measurement in papermaking.

Stiffness was measured by vigorously shaking a piece of paper and listening to the noise. In fact, so-called bond paper was quality assessed by its noise level! It had to be very noisy!

# EARLY PAPER TECHNOLOGY

Paper stiffness was probably the first paper property which interested me. It was easy to note that deflection of a sheet of paper was time dependent. The existing stiffness instruments all gave different values, because of the various times needed to make the measurement and of course the relative humidity in the room. It was obvious that paper was not a simple elastic material.

Professor W. Brecht in Darmstadt had told me that paper researchers had to make two contributions, a sizing meter and a stiffness meter. I never had time to design a sizing test.

I built my stiffness tester from a loudspeaker connected to a frequency generator<sup>2</sup>. On the paper cone the paper strip was fastened and the frequency varied until maximum deflection at the quarter wavelength of the paper sample. Cutting strips radially, I could demonstrate in a polar diagram the variation in "true" elasticity

module in the sheet. This was possibly the first paper testing made outside the CD and MD direction.

Attempts to isolate a plastic component in the material did not succeed.

Instead I turned to stress - strain experiments. My instrument design rules required the ability to study the stress-strain behaviour at different speeds and at different relative humidities. I needed to measure elongation far more in detail than before. I also wanted to be able to study the behaviour when reversing the load. I choose to work with constant rate of elongation and consequently to study primarily stress relaxation. The reason was that relaxation was theoretically simpler than creep studies.

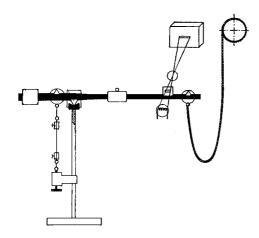


Figure 1. First stress-strain instrument.

The exploring instrument was built on a modified Stathmos bathroom balance. Stathmos was famous for their hard steel wedges. Initially, the instrument was built as a chain balance, using a bicycle chain for loading<sup>3</sup>. The paper strip was elongated by a hand-driven micrometer, the constant rate obtained by listening to a Metronome. The needed instrument three laboratory assistants in the dark room: One turning the elongation screw, a second letting out or pulling in the bicycle chain to reach balance, and a third to note the data, from which the stress-strain curve was drawn, Figure 1.

Relative humidity was upheld by enclosing the paper strip and the two paper clamps in a thin latex tube of the mark "Ramses". Air conditioning was provided by leading air from a series of salt solutions into this envelope.

The interesting results from this instrument motivated mechanization, which was essentially carried out by Josef Kubat, then laboratory assistant and later a wellknown rheologist. New types of servomotors and excellent shop work by Sune Holm helped him.

We had now the possibility to study paper making in a detail not possible or even attempted at the time<sup>4</sup>.

In those days paper machines were stopped on Saturday afternoon and restarted after housekeeping on Monday morning. This gave us an opportunity to carry out full-scale experiments late on Saturdays. The Norwegian mills were far more willing to let us do such experiments than the Swedish, and several Norwegian mills sent us guest workers. We preferred to experiment on machines making strong un-filled papers from one single chemical pulp quality.

We added dyed fibres to study fibre orientation. We took samples at all open draws in the paper machine - and they were then many – and dried the samples to study the kinetics of papermaking. This analysis was always made in all directions and many positions across the paper machines. It is interesting to note that such experiments today would be almost impossible. Modern paper machines run continuously and are up to five times wider and ten times faster than the machines we studied.

It was found that many paper properties effects depended on the draws of the paper in the machine direction and the shrinkage in the cross direction. Splitting the paper lengthwise in the paper machine already after the couch and pressing and drying the two endless parallel sheets separately gave important new information. Paper curl could be studied by comparing fibre orientation on wire and felt side.

Early on it was obvious that our results could be expressed in terms of viscoelasticity. We published seven articles under the general title "Paper as a visco-elastic body"<sup>3</sup>. Writing for papermakers I tried to avoid fancy words, new to them. The term "Rheology of Paper" was first used as a title of a symposium in London in March 1949 when Dr G.W. Scott Blair participated in the discussion<sup>5</sup>.

This was a period when mechanical models of plastic material were popular. I was influenced by H. Leaderman at Bureau of Standards who had worked on textile materials and had bought a copy of "Mechanical behaviour of high polymers" by Turner Alfrey Jr, reeking with mechanical models.

I had found that a three-element model was reasonably applicable to paper. H. Eyring had advanced a non-Newtonian rheological model where holes were moving opposite to the strain. This model had 1945 been applied to wool fibres by G.Halsey et al.<sup>6</sup>.

Bertil Ivarsson and I believed that this model could be usefully applied to practical paper testing<sup>7</sup>. The provision was to find a fast method to evaluate stress strain curves. We first found that the published mathematics had serious errors. Once corrected we did not succeed in solving the problems necessary to have the method accepted in practise. Even using simplified master curves and some training it took 15 minutes to determine the elastic modulii and a value of the apparent non-Newtonian viscosity.

Parallel with these theoretical works new instruments were designed. Kubat, Nyborg and Steenberg<sup>7</sup> described an instrument for study of the response of paper to low frequency sinusoidal strain. This study was based on the observation that paper showed pronounced strain hardening. Repeated

strain during handling of cement bags might have an effect on paper structure. Repeated loading would eventually result in fatigue and a brittle break.

## HIGH SPEED TESTS

We now turned our attention to higher rate of elongation. Paper testing method applied the load at very low rates, unrealistic to describe the paper behaviour, say, when big cement paper bags were dropped. The speed of the STFI tester could only be increased ten times, still far to slow.

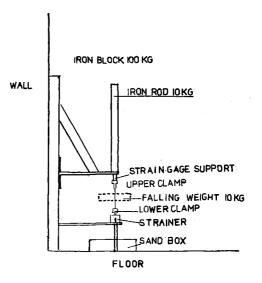


Figure 2. Fast stress strain instrument.

An instrument was built were freely falling weight produced high and controllable rates of strain, Figure 2. The vertically oriented paper test strip was fastened in two clamps. The lower of this was hit by the falling weight, straining the paper to the breaking point. The upper clamp had an extremely thin tube extension around which strain gauges were glued<sup>4</sup>. The design had to have minimum inertia. It was turned from the root part of a dour-aluminium DC 3 propeller, Figure 3. The stress strain curve was recorded on a 1947 model oscillograph. To get sufficient light for photographing the stress strain curve on the small cathode ray screen the oscillogram had to be very tiny.

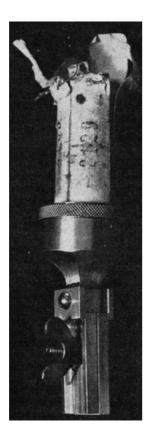


Figure 3. Fast strain gauges on Al-tube.

The film was copied enlarged and the stress strain curve evaluated.

Number five in the Series "Paper as a visco-elastic body" was the first that dealt Andersson<sup>8</sup> with impact conditions. described the propagation of a tension or compression wave in a non-Hookean Above a critical velocity the material. sample ruptured at the impact end, with the inertia of the strip acting as a virtual second clamp. The critical velocity could be shown to have a physical meaning. An instrument was built to determine its value. A heavy metal wheel was accelerated to a specific periphery velocity. It then engaged a paper clamp attached to a free paper strip. The lowest speed that broke the paper at the clamped end was taken as the critical velocity. For newsprint this velocity turned out to be about 23 m/sec. The test needed a new clamp for each test and rather few tests were made. It had a high spectator value.

The group consisting of Olle Andersson and Bertil Ivarsson and the author received many invitations to speak and guest workers arrived from Norway, Canada, England and USA. Among them who published contributions from this time can be mentioned Toby Rance, Wiggins Teape Co and Alfred Nissan, Bowater, UK, later Westvaco, US<sup>4</sup>. Both eventually were honoured with the TAPPI gold medal.

## FROM LABORATORY TO FACTORY

The rheological results had practical consequences. Not only did they increase the understanding of the paper machine operations and thus formed the basis for new control measures. It also resulted in new types of paper. Two examples will be given.

Newspapers were at his time printed from a hemi-cylindrical metal cast made from the original flat set Gutenberg letters and clichés. On the composed page set a heavy moist paper board, a "flong", was pressed. The board got an imprint of the page. After drying it was bent in a hemi circle and put into a casting machine. Two hemi-cylindrical casts were put together and formed the print roll.

We designed a method to produce flongs, which saved paper for the newspapers. This was based on our ability to control not only the amount of shrinkage but also its direction. A flong which shrunk in the cross direction but not in the machine direction allowed the printer to use his old types, but sell a printed paper where the letters in the cross direction were slightly thinner. Paper was saved by running narrower roll of newsprint. Bertil Ivarsson, mv first assistant, introduced the method at Beveridge Paper Mill in Indianapolis, US and they held the American market.

Multiwall bag paper was a very large market before the advent of plastic foils. We had realised that the area under the stressstrain curve (the work of breakage) was much more important than the tensile strength. The paper maker knew how to reach high tensile strength paper but not how at the same time produce stretch, a property which could not even be reliably measured at the time.

We had found that by allowing the paper to shrink freely we could increase stretch, and thus the work of breakage. I coined this "micro-creping", a word later used a trade name for the quality<sup>9</sup>.

The technical development was lead by Sven Spangenberg at Fiskeby and later Korsnäs mills. Initially air was blown between the sheet and the drying cans to allow free drying. Later Svenska Fläkt Fabriken AB developed air-dryer together with Claes Allander, later professor in heat technology at the Chemical Faculty, KTH).

West Virginia Pulp and Paper Co was the first American company to buy the commercial model of the STFI-tester, later called the Alwetron. There under the guidance of Alfred Nissan they developed the high stretch "Clupac" multi-wall bag paper, which become a world standard.

The new stronger Swedish multiwall bags could be made with fewer sheets of higher basis weight than before, but only from never dried kraft pulp. It rapidly became standard and the continental paper makers, using dried pulp, had to leave the market. The diminishing international demand for unbleached softwood kraft forced some Swedish mills to install pulp-bleaching units.

In the early 1950-ties complementary studies of paper rheology started at many places. Textbooks<sup>10</sup> quoted our early work expensively. The law of decreasing returns

had set in and I tried to leave the field for new ventures. Fibre suspensions have their own rheology<sup>11</sup>.

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