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DISTRIBUTION OF ENERGY CONSUMPTION DURING THE STRAINING OF PAPER

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Synopsis The thermodynamic behaviour (that is, the simultaneous mechanical and thermal behaviour) of paper and other sheet-like materials will be reported. It will be shown that the thermodynamic analysis will reveal much more of the deformation mechanism than the mechanical analysis alone.

Results obtained show that the initial straining of paper is controlled by energy elastic forces and in accordance with Kelvin's thermoelastic equation. The plastic region straining of paper is controlled by irreversible intrafibre deformation. Simultaneously, some interfibre bond breakage does occur, but this breakage is partial and it is not a prerequisite of plastic deformation of paper.

Introduction

DURING the past 10–15 years, the structure and structural behaviour of paper has been studied quite intensively. The results of many of these studies have been reported or reviewed in the transactions of the three fundamental research symposiums organised by the British Technical Section.[1–3] Simultaneously, along with the increase in the understanding of the structure of paper, many theories have been put forward to relate the mechanical behaviour of paper to the mechanical behaviour of the component fibres.

General

Paper is almost a unique material, because its principal structure components fibres are of macroscopic size and heterogeneously arranged. In addition, fibres in a typical sheet of paper contain heterogeneities at various levels of the structure of cell wall (Table 1). The way fibres are bonded to each other (that is, formation) is also very heterogeneous. (In this connection, it is good to remember that in a typical sheet of paper there is more material in the bonded segments than in the free segments.)

Under the chairmanship of Prof. H. W. Giertz

TABLE 1—HETEROGENEOUS STRUCTURAL FEATURES OF FIBRES IN A 'TYPICAL' PAPER

Variations in physical parameters of fibres
Distribution of chemical components in cell wall
Distribution of length of free and bonded segments
Extent of Page & Tydeman effect in bonded segments
Extent of Jentzen effect in free segments
Amount of misalignment zones
Amount of lumen bonding
Bonding geometry of delaminated cell wall
Possible longitudinal contraction of the elementary fibrils caused by pulping

Based on the great amount of heterogeneity in a typical sheet of paper, one might think that such a complex body does not have well-defined, regular mechanical properties, but this is not the case. For instance, the load/elongation curve of a typical paper shows similar features to the load/elongation curves of other planar materials like plastics and metals.

The combined results of the research on individual woodpulp fibres [4–10] show the same principal mechanical behaviour for woodpulp fibres as for paper.

It is also generally accepted that similar bonding geometries and bonding forces are involved between the larger structural components of the cell wall as are involved in interfibre bonds of typical papers. An important question therefore is whether the viscoelastic and apparent plastic behaviour of paper is the result of a significant intrafibre phenomena or whether this behaviour is mainly caused by an interfibre phenomena—that is, by the prerupture breakage of fibre-to-fibre bonds.* Such structurally different papers as (*a*) filter paper, (*b*) 'typical' paper and (*c*) glassine and condenser papers show similar load/elongation curves (Table 2). Thus, it has proved to be difficult to answer the above question.

TABLE 2—CHARACTERISATION OF VARIOUS GRADES OF PAPER

Type	*Example*	*Characteristic features*
Porous papers	Filter papers	Low RBA, relatively long free segments
Typical papers	Bond, sack, publication grades, packaging papers	Moderate RBA, length of free segments less than the collapsed width of fibre
Dense papers	Glassine, condenser paper	High RBA, shape of individual fibres lost, practically no free segments

Structural behaviour during plastic region straining

Three different mechanisms have been put forward as hypotheses for the plastic region of the load/elongation curve of paper. These mechanisms are summarised in Table 3. It should be noted that the division into these three

* With bond breakage, it is understood here that part of the fibre-to-fibre bond or the whole bond breaks irreversibly

classes has been made somewhat arbitrarily. For instance, Van den Akker[26] has emphasised that besides the breakage of fibre-to-fibre bonds one should not lose sight of the simultaneous intrafibre creep and stress relaxation. A similar statement has been made by Sanborn.[36]

TABLE 3—MECHANISMS RESPONSIBLE FOR PLASTIC REGION DURING STRAINING OF PAPER

Mechanism	*Used by*
1. Viscoelastic behaviour	Steenberg & co-workers,[12, 13] Andersson,[14] Kubát & co-workers,[15–18] Sternstein & Nissan[19]
2. Prerupture breakage of fibre-to-fibre bonds	Rance,[20, 21] Craven,[22, 23] Nordman & co-workers,[24] Ranger & Hopkins,[25] Van den Akker,[26] Kallmes & Perez,[27] Perez[28]
3. Matrix model	Page & Tydeman,[29] Helle,[30] Giertz,[31, 32] Page,[33] Wrist,[34] Houen[35]

It is important to note that in the famous studies of the viscoelastic mechanism by the Swedish workers, the discussion of the results and their implications on the structure and deformation of paper have not required the introduction of interfibre bond breakage as an important part of the plastic deformation.

The term matrix model has been introduced by the Norwegian school of paper scientists.[32, 35] According to the matrix model, the basic deformation mechanism contributing to the plastic region straining of paper is the irreversible shear deformation. This shear deformation takes place between the amorphous matrix and the microfibrils in those bonded segments of the cell wall that have been made active by the consolidation* or by the matrix breakage of earlier active segments. Fibre-to-fibre bond breakages occur (partial or total) when the local stress concentration exceeds the load-carrying capacity of the amorphous matrix in the bond region, which has already been broken down by the removal of the microcompressions. Thus, the breakage of fibre-to-fibre bonds is a natural consequence of the plastic deformation of the cell wall in the bonded segments, not a prerequisite of the plastic deformation. According to the matrix model, permanent set is caused by the breakage of the amorphous matrix, because the microfibrils of the bonded segments do not return to their compressed positions after the removal of the external load.

Observation of interfibre bond breakage due to straining

Most of the experimental results indicating prerupture bond breakage are indirect. They are based mainly on optical data. Rance related the bond breakage to the opacification of paper following the plastic region straining.[37]

*Following from the Page & Tydeman effect[29]

Similar results have been reported by Ranger & Hopkins.[25] It is now generally agreed that the increase of the scattering coefficient during plastic region straining is related to the bond breakage.[24, 36, 38, 39] Similar results have been obtained also using the gas sorption technique.[40–43] It should be remembered, however, that the light scattering technique measures changes in surfaces that are not necessarily in molecular contact and that the gas sorption technique measures changes also in surfaces that may not be involved in the fibre-to-fibre bonding.

Page & Tydeman have observed by light microscope the breakage of bonds due to straining.[44, 45] These results of Page & Tydeman indicate that most of the bond ruptures are partial. In this connection, one should keep in mind that the results were obtained with a technique that measures changes in surfaces that are not necessarily in molecular contact. One should also note that the results were obtained by analysing surface fibres—that is, fibres having a different bonding geometry than fibres in the bulk of the sheet.

There are also other indirect observations that have been related to the breakage of the interfibre bonds.[25, 36, 46, 47] On the other hand, indirect experimental results have been reported that do not support the significant occurrence of prerupture interfibre bond breakage.[48–50]

Based on the review of the literature, it seems that no conclusive experimental evidence exists for the significant occurrence of the complete breakage of interfibre bonds during the straining of paper. Neither has it been experimentally verified that the breakage of fibre-to-fibre bonds would cause the plastic region in the load/elongation curve of paper.

Experimental

RESULTS will now be presented of a thermodynamic examination of the load/elongation behaviour of paper.[51] The objectives of this investigation were to determine how the different structural features of paper affect its thermodynamic behaviour. Results from these experiments and the corresponding data from other planar materials were analysed to establish the most significant deformation mechanism during the straining of paper. Additional information relating to the roles of interfibre and intrafibre deformation was gathered from stress relaxation data, from the measurements of straining induced changes in the light scattering coefficient and from the analysis of fibre behaviour in the rupture zone of the tensile specimens. The microcalorimeter, specially constructed for this work, has been described elsewhere.[52]

The general thermodynamic aspects of deformation are described in appendix 1. It can be concluded that a great deal more may be learned about the deformation when the load/elongation curve and the heat effects are

recorded simultaneously. For instance, the thermodynamic concepts of deformation can be applied to Nordman's bonding strength concept (NBS value).[24] If all the irrecoverable work of a load/elongation cycle would go into the creation of unbonded surface area, then there should be no net heat phenomena associated with the load/elongation cycle.* On the other hand, if considerable irreversible heat generation is obtained during the straining of paper, then one has to conclude that an appreciable amount of the work of straining is lost in the irreversible structural changes of the cell wall.

Material tested

Because of the operational principle of the microcalorimeter, paper and other hygroscopic specimens had to be tested in a dry condition. Except for two instances, all the straining and destraining was carried out in a dry nitrogen atmosphere with the microcalorimeter. The elongation interval studied was between 0 and 5·0 per cent. Table 4 lists the various sheet-like materials tested during this work. The materials tested represent three categories—

1. Conventional paper having filaments of a very heterogeneous structure.
2. Synthetic paper being composed of axially homogeneous filaments.
3. Films and foils that do not have the filament structure.

In what follows, results will be described mainly for paper (natural and synthetic), regenerated cellulose film and aluminium foil.

TABLE 4—MATERIALS TESTED

Materials	*No. of specimens*
100 per cent rag paper, cross-direction	31
100 per cent rag paper, machine-direction	17
Blotting paper	1
High-stretch sack paper (Clupak)	1
Bleached kraft handsheet (4 qualities)	10
Cellulose acetate	1
Cellulose film (4 qualities)	23
Teflon (4 qualities)	4
Polythene* (3 qualities)	5
Other plastic films (2 qualities)	2
Metal foils (3 qualities)	10
Metal filaments (2 qualities)	2
Rubber	1
100 per cent rag paper†	1
100 per cent rag paper‡	1

* Includes synthetic paper
† At 11 per cent rh
‡ At 48·6 per cent rh

* In order to be exactly valid, the Nordman bonding strength concept requires that the work lost in the load/elongation cycle is not used to generate new residual strain energy

RESULTS AND DISCUSSION

Thermodynamic behaviour of various sheet-like materials

Machine-made paper

FIG. 1 gives the mechanical and thermal response of a slack-sized 100 per cent rag paper (79 g/m²) to straining in the cross-direction. Similar results for the machine-direction specimen are shown in Fig. 2.

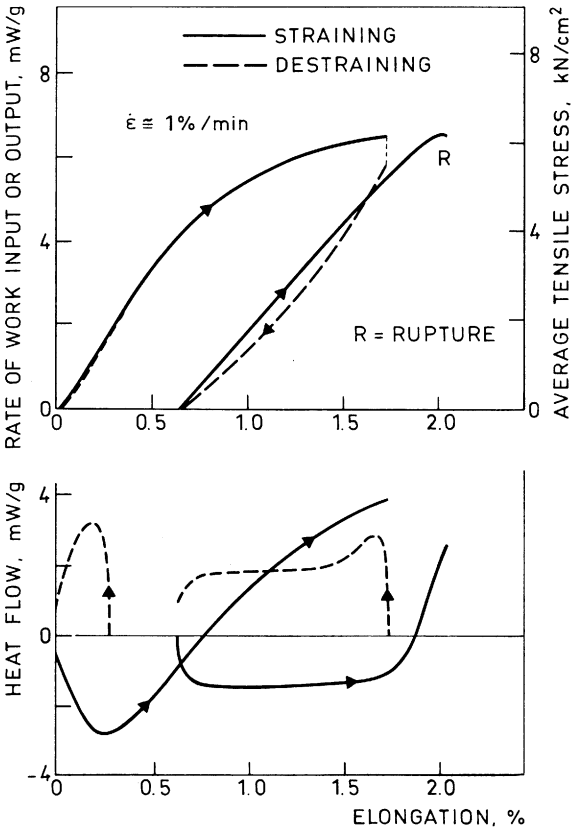


Fig. 1—Thermodynamic behaviour of 100 per cent rag paper MD

The average tensile stresses were calculated using the solid cross-section of the specimen. It should be noted that the heat flow curve in reality does not end sharply when either the straining or destraining ends. Instead, there is a certain decay of the heat flow to the zero value. This decay may take quite

some time in the case of strong viscoelastic phenomenon (see stress relaxation in Fig. 2). Generally, the decay of heat flow is not shown in the graphs, because the heat flow is given as a function of elongation. One should also note the sign convention; a negative heat flow means that the specimen is receiving heat from the surroundings and a positive heat flow means that the specimen is generating heat. The rate of work input coincides with the straining part of the load/elongation curve. The rate of work done by the specimen during destraining coincides with the destraining part of the load/elongation curve.

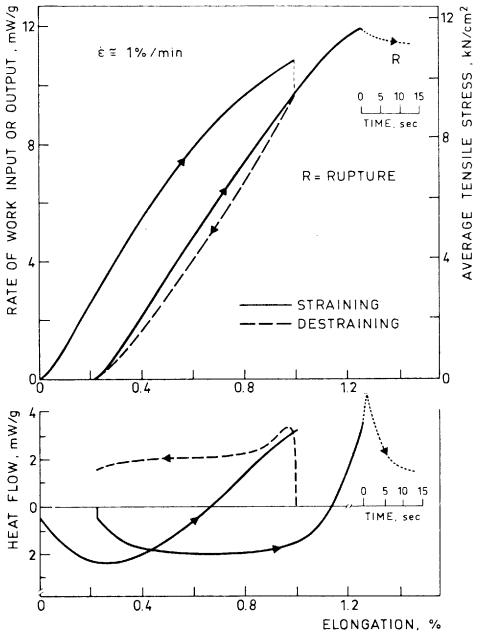


Fig. 2—Thermodynamic behaviour of 100 per cent rag paper MD

From Fig. 1, one sees that the specimen tends to cool during the initial straightline response to straining—that is, it is receiving heat from the surroundings. If destraining is carried out from this stage of the straining, the load/elongation curve shows practically no irreversible phenomenon. Thus, heat is flowing out of the specimen during the destraining in accordance with a reversible phenomenon.

If one continues the straining beyond the apparently Hookean response, the rate of load development slows down and the rate of heat flow to the specimen

changes gradually to a rate of heat generation.This heat generation is an irreversible phenomenon, because heat is given out throughout destraining. One should also note that a considerable amount of work is lost irreversibly in the dissipative heat generation. In other words, the apparent plastic region straining of paper is a true plastic deformation—that is, to a mechanically and thermodynamically irreversible deformation (see Fig. 13, appendix 1).

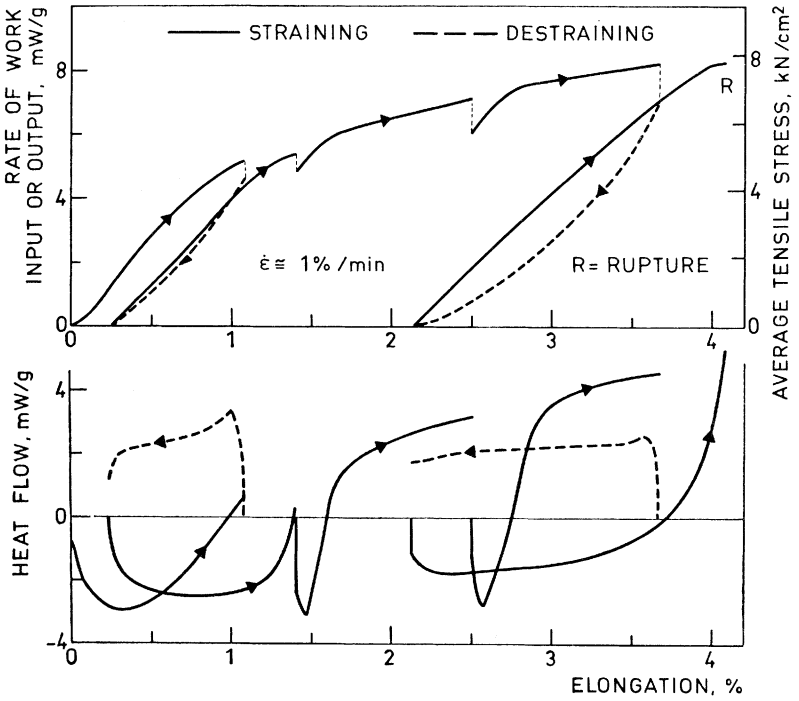


Fig. 3—Thermodynamic behaviour of clupak sackpaper MD

When a mechanically conditioned specimen is strained, it exhibits an apparently Hookean behaviour over a longer straining interval than does the original specimen. At the same time, the interval of receiving heat from the surroundings is also longer. Again, when the apparently straightline response is exceeded (end of mechanical conditioning), the specimen quickly starts to generate heat once more. (One should note that the mechanical conditioning has lowered the slope of the load/elongation curve.)

The reported thermal phenomenon is qualitatively similar to that obtained by Tydeman[53] for heavily refined sulphite paper and by Joule[54] for dry wood.

High-stretch sack paper

The thermodynamic behaviour of Clupak sack paper (72 g/m^2) in the machine-direction is shown in Fig. 3. The basic thermal phenomenon is similar to that of 100 per cent rag paper—

1. During the initial, apparent elastic region straining, heat is absorbed by the specimen.
2. During the plastic region straining, heat is irreversibly generated by the specimen.
3. If straining is stopped during the plastic region elongation and the initial stress relaxation is allowed to take place, continuation of straining causes a momentary heat absorption followed by a quick reversal to heat generation as soon as the previous stress level is reached and the apparent elastic response ends. The rate of heat generation during the continuation of plastic region straining follows the path one would have predicted by extrapolation of the previous heat flow curve.
4. During destraining, heat is given out.
5. After a load/elongation cycle involving plastic region straining, the length of the initial heat absorption interval increases with a simultaneous increase in the apparent straightline response to straining.

Comparison of the last destraining-straining stress curves shows that a moderate element of internal friction is present. This can also be concluded from the corresponding heat flow values—that is, the rate of heat given out is higher than the rate of heat absorption (respectively $-1{\cdot}6$ mW/g compared with 2·2 mW/g at an elongation of 2·5–3 per cent).

Blotting paper

The blotting paper studied was the same as that used in TAPPI method T205 m-58. Its grammage was 260 g/m^2 and solid fraction about 0·3. The thermodynamic behaviour of this paper in the machine-direction was very similar to that of the rag paper in the cross-direction (Fig. 1).

Handsheets

It is well known that refining, wet pressing and drying tension greatly affect the structure of paper and, consequently, the mechanical properties of paper. The effect of these variables on the thermodynamic behaviour of paper was studied also in this investigation. Time does not allow a full account to be given; an interested reader is referred to the original work.[51]

The sheetmaking conditions are listed in Table 5. The relevant papermaking properties are given in Table 6. The general thermodynamic behaviour of the handsheet was similar to that of machine-made papers—that is, during the

apparent elastic deformation, the thermal phenomena were reversible and, during the apparent plastic straining, an irreversible heat generation took place (Fig. 4).

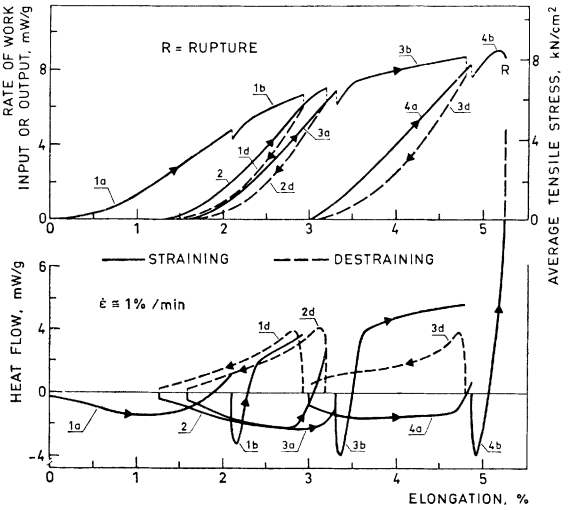


Fig. 4—Thermodynamic behaviour of handsheet D

Because of the free drying, the handsheets had a slightly cockled and creped appearance. This was especially true for handsheets made from refined pulp.

TABLE 5—HANDSHEET DATA FOR BLEACHED KRAFT SOFTWOOD PULP

Specimen	*No. of specimens*
Handsheet A (free drying,* 650° CSF)	3
Handsheet B (free drying, 93° CSF)	3
Handsheet C (ring drying,† 93° CSF)	3
Handsheet D (no wet pressing, free drying, 93° CSF)	1

* On purified sand bath, 96 per cent rh
† 6 per cent rh during ring drying

The cockled appearance resulted in a noticeable toe to the load/elongation curve, indicating that the slope of the curve was increasing during the apparent elastic straining. Simultaneously, the rate of heat absorption was increasing. This result differs from the behaviour of machine-made papers, for which the elastic response was accompanied with a relatively straightline increase in load and a relatively constant rate of heat absorption (Fig. 1–3). This result of strain hardening and increased rate of heat absorption seems clearly to

indicate that the amount of material sharing the load is increasing during the initial stage of straining. Actually, this is to be expected, because those segments that dry last in a freely dried sheet will form bonds and microcompressions last and, in so doing, will introduce some slack into the already dried structure.[22, 23, 29] During straining, however, the segments and interfibre bond sites that dried last will respond first. Only after some straining will those fibre segments that dried earlier (in the beginning of consolidation) start sharing the load.

TABLE 6—HANDSHEET PROPERTIES IN DRY NITROGEN

Handsheet code	*Tensile strength, kN/cm²*	*Breaking elongation, per cent*	*Specific scattering coefficient, cm²/g*	*Change in specific scattering coefficient due to straining to rupture, cm²/g*
A	8·0	3·2	320	24
	(7·6)	(5·4)	n.d.	n.d.
B	9·2	5·3	152	41
B_m*	(10·9)	(14·0)	74	130
C	14·8	2·0	197	22
D	8·4	5·2	171	37
High-stretch	7·8	4·1	—	16
sack paper, MD	(9·1)	(8·0)	n.d.	n.d.

Numbers in parenthesis are for testing at 50 per cent rh
n.d. = not determined
* This specimen was indentical to handsheet B, except that the degree of refining was 60° CSF is compared with 90° CSF for handsheet B

Compared with the freely dried handsheets (breaking elongation in dry nitrogen 3–5 per cent), the ring-dried handsheet (breaking elongation 2 per cent) did not show any noticeable strain hardening. Its thermodynamic behaviour resembles that of the machine-made rag paper (Fig. 2).

Magnitude of dissipative heat generation

The relative amount of heat generation for various handsheets is listed in Table 7. The corresponding data for other paper specimens are also included. It can be concluded that the rate of heat generation generally during the plastic region straining is 40–60 per cent of the corresponding rate of work input. Simultaneously, the apparent relative rate of creep varies from 0·65 to 0·85, that is, the load is increasing with a rate of only one third to one sixth of the initial slope of the load/elongation curve.

It is important to note that, although the structural features of these papers are different and although the mechanical properties vary considerably, the relative magnitude of irreversible heat generation is fairly constant.

TABLE 7—DISSIPATIVE HEAT GENERATION IN VARIOUS PAPER SAMPLES

Specimen	*Breaking elongation* ε_{max}, *per cent*	*Observation elongation* ε, *per cent*	*Ratio of rate of heat generation to the corresponding rate of work input,* $\dot{q}\downarrow/\dot{w}\uparrow$, *per cent*	*Apparent relative rate of creep,* u/v
A	3·2	2·0–2·5	50	0·75
		3·0	75	0·9
B	5·3	4·0–5·0	60–65	0·8–0·85
C	2·0	1·5	45–50	0·8–0·85
D	5·2	2·8	50	0·55
		4·8	65	0·85
Rag paper, MD	1·2	1·0–1·1	35	0·65–0·75
Rag paper, CD	1·9	1·5–1·8	60	0·8–0·85
Sack paper, MD	4·1	2·4–3·6	45–55	0·8–0·85
Blotting paper	1·8	1·4	50	0·85

Relative rate of creep $= 1-(df/d\varepsilon)/(df/d\varepsilon)_{\varepsilon=0}$
Rate of straining was about 1 per cent/min

As has already been mentioned, the dissipative heat phenomenon can be removed by mechanical conditioning. In other words, it is possible to strain the mechanically conditioned specimen to rupture in such a manner that the specimen is absorbing heat all the time and exhibiting very little plastic region yielding.

The following conclusions can now be made—

1. Plastic region straining of paper involves irreversible structural changes.
2. Viscoelastic response and especially the so-called entropy force do not account for the plastic region deformation of paper.
3. Classical frictional forces (interfibre or intrafibre) do not play a significant role in the plastic region straining of paper.

Synthetic papers

Two varieties of synthetic paper were used. One represented a well bonded paper-like fibrous structure (Tyvek 1058 spunbonded polyolefin paper, 54 g/m²); the other paper represented a porous fibrous structure (Zitex fluorocarbon paper, 92 g/m²).

Fig. 5 shows the thermodynamic behaviour of the spunbonded polyolefin paper. It can be seen that the load/elongation curve behaviour resembles that of high-stretch sack paper (Fig. 3) and that the specimen is absorbing heat all the time during straining. There is no clear yield point as was the case with some papers. Anyhow, there is clear evidence of some kind of yielding during the latter part of extension (apparent relative rate of creep = 0·7), yet no evidence of large-scale irreversible heat generation is observed. Although there is a considerable hysteresis loop formed in the load/elongation cycle, the net heat of the cycle is negative. This means that no significant irreversible

structural changes have taken place. Furthermore, the formation of the hysteresis loop and the apparent plastic behaviour of this spunbonded polyolefin paper seem to be a viscoelastic phenomenon. One should note that this explanation is applicable only to the very short-range extension of this specimen.

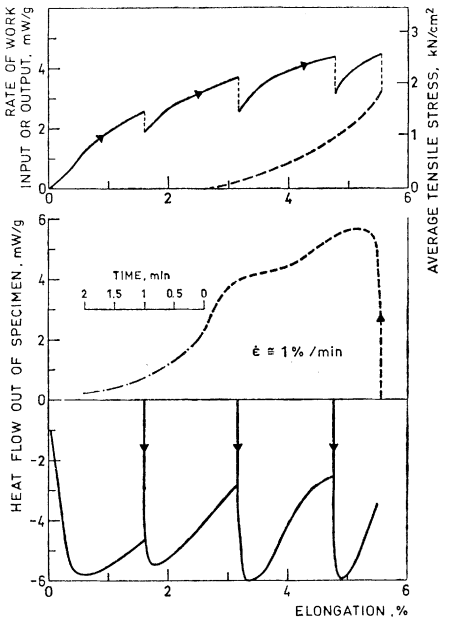


Fig. 5—Thermodynamic behaviour of spunbonded polyolefin paper

The thermodynamic behaviour of the porous fluorocarbon paper was similar to that of the spunbonded polyolefin paper. After the initial straightline response, the rate of heat absorption decreased sharply, but it did not change to a positive value even after considerable apparent plastic deformation.

Regenerated cellulose film

Fig. 6 shows the thermodynamic behaviour of a plasticised cellulose film specimen (50 g/m²) in the cross-direction. It is seen that the initial heat flow is similar to that with papers, although the value of heat flow per unit mass is higher. It is also seen that the general thermodynamic behaviour is quite similar to that of synthetic papers. In addition, with cellulose film, there is no evidence of large-scale irreversible heat generation during the latter part of

the straining even though there is a noticeable amount of yielding in the load/elongation curve. (Apparent relative creep rate varies 0·6–0·8 in the elongation interval from 3 to 4 per cent during the last run in Fig. 6.)

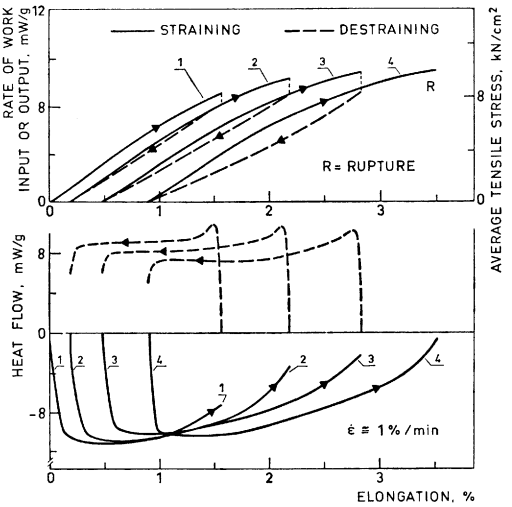


Fig. 6—Thermodynamic behaviour of plasticised regenerated cellulose film CD during successive load/elongation cycles

It seems that the gradual (and moderate) yielding of cellulose film is connected to a viscoelastic phenomenon, not to an irreversible structural change as was the case with paper.

Aluminium foil

The thermodynamic behaviour of aluminium foil (110 g/m²) in the machine-direction is illustrated in Fig. 7. (The stress was calculated per solid cross-section using the density value of 2·7 g/cm³ and the weight of the specimen.) It is seen that during the initial Hookean region heat is flowing to the specimen. This heat flow reaches a maximum before any yielding is observed in the load/elongation curve. When the yield transition period is completed and the plastic deformation starts, the heat flow changes to a dissipative heat generation. During the plastic region straining, the apparent relative creep rate is 0·98 and the rate of heat generation is 50 per cent of the rate of work input. The latter value is in accordance with similar data in the literature.[55, 56]

During destraining, heat is always evolved. Practically no hysteresis loop is formed between the first destraining and the second straining, indicating a reversible deformation. Besides, the heat flows of destraining and straining

are mirror images of each other, just as one would expect for a reversible deformation. When the stress level of the previous straining is reached, however, a quick change to plastic straining and irreversible heat generation takes place. Simultaneously, there is a sharp change in the slope of the load/elongation curve.

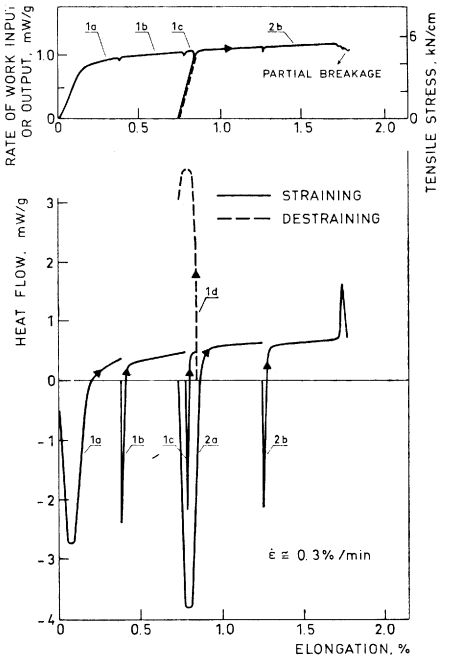


Fig. 7—Thermodynamic behaviour of aluminium foil MD

The continuation of straining after an intermediate stress relaxation pause causes an initial short heat absorption, then the dissipative heat generation starts just as in the case of papers made from wood fibres.

It is generally accepted that the plastic deformation of aluminium and other metals occurs by slip (intergrain and intragrain 'plastic flow'). This slip can be visualised as irreversible shear deformation between the structural units of aluminium.

Origin of thermal phenomenon during deformation

THE tendency of a tensile specimen to cool during apparently elastic straining and to warm up during destraining is a universal phenomenon for all those materials that have a positive thermal expansion coefficient and a Poisson

ratio of less than 0·5. This type of heat phenomenon of uniaxial deformation is called Kelvin's thermoelastic effect. Lord Kelvin derived the thermoelastic equation by the means of classical thermodynamics some 120 years ago. The pertinent features of Kelvin's thermoelastic equation are given in appendix 2.

One can also derive the thermoelastic equation from molecular considerations. The thermal phenomenon during elastic deformation is connected to changes in the vibrational states of the atoms.

Kelvin's thermoelastic equation provides one possible explanation of the connection observed between the beginning of plastic deformation and the beginning of irreversible heat generation. Usually, it is assumed that the beginning of plastic response to deformation is connected with structural breakdown. Based on this assumption, one can account for at least part of the irreversible heat generation by the release of heat of straining and of stored strain energy by those structural elements involved in the internal rupturing. If it is further assumed that some rebonding is possible, then one can account for the fact observed with many paper specimens that heat generation during destraining is independent of the straining-induced microrupturing. This is because those elements that undergo destraining generate heat; those elements that after rebonding undergo compressive loading also generate heat. It should be emphasised that this concept of microrupturing is not meant to be used quantitatively as a new theory of plastic deformation of natural papers.

Short-term stress relaxation

FIG. 8 gives the refined stress relaxation data for rag paper, cellulose film and aluminium foil. The stress relaxation periods varied 2–3 min. The data has been fitted to the stress relaxation equation used by Craven[23] and by Johanson & Kubát[57]—that is, $\sigma_0 = \sigma_0 - b \log t$. Here, σ_0 is the 'unrelaxed' stress and b is a constant. The straightline region of relaxation was reached in 7–10 s in this study, except for cellulose film, for which about 30 s were needed. A reasonably good straight line could be fitted for the experimental data. (No attempts were made to check the applicability of the 'straightline' relationship over longer periods than 2–3 min of time.)

It can be concluded from results in Fig. 8 that, besides the greatly different structures of the various specimens, they all seem to have approximately the same type of mechanism for the logarithmic stress relaxation. It is important to note that the results obtained with the paper specimens indicate that the logarithmic stress relaxation is governed by the same mechanism for both the apparent elastic region and for the plastic region.

The initial stress relaxation in paper specimens was faster than in cellulose film and the initial relaxation of aluminium foil also was faster than that of

cellulose film. It is considered therefore that the results obtained do not support the concept that the relaxation phenomenon in paper for lower stresses is due to the viscoelasticity and, for higher stresses, mainly to the breakage of fibre-to-fibre bonds. This holds even for loads very close to the ultimate tensile strength of the paper specimens. Similar structural phenomena control the relaxation in both regions of the load/elongation curve of paper.

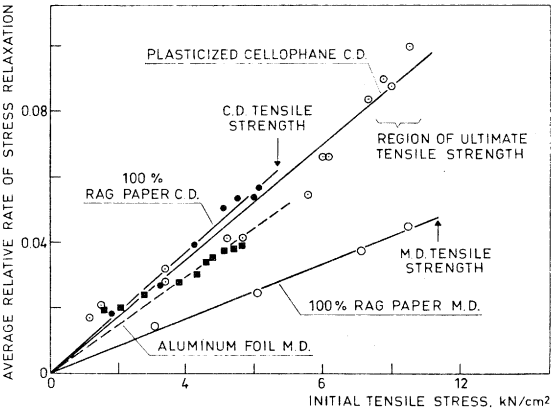


Fig. 8—Refined stress relaxation data for rag paper, regenerated cellulose film and aluminium

Apparent linear thermal expansion coefficient

ONE can use Kelvin's thermoelastic equation—appendix 2, equation (*4*)—to calculate the linear thermal expansion coefficient of various materials, provided the uniaxial deformation (from which the data is collected) is elastic. In this study, the destraining data was used on the assumption that the irreversible phenomenon in destraining was of a small magnitude.

Fig. 9 shows that straight lines through the origin were obtained in accordance with Kelvin's thermoelastic equation when the destraining data of the handsheets were plotted. Similar good fits were obtained with the other paper specimens and with the various sheet-like materials used in this study.

Table 8 lists the calculated linear thermal expansion coefficients for various papers. This table also contains the corresponding average initial destraining modulus. Two points should be noted—

1. The calculated values of α are very close to the measured values reported by Kubát, Martin-Löf & Söremark,[58] who carried out the measurements in dry nitrogen.
2. There is an approximate inverse relationship between the thermal expansion coefficient and the destraining modulus of the same paper. This relationship was observed by Kubát and co-workers even for the straining modulus.

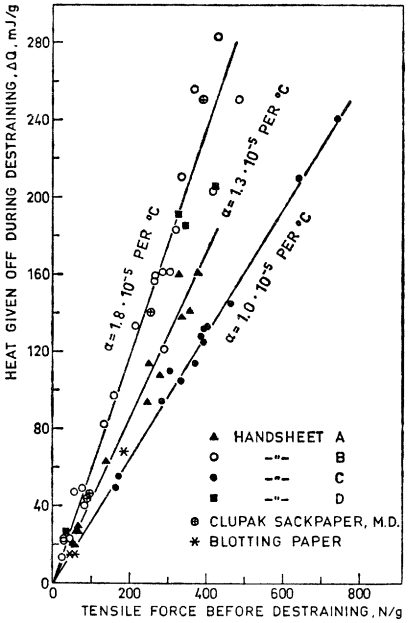


Fig. 9—Relationship between heat output and tensile force in destraining runs of different handsheets and miscellaneous papers

TABLE 8—APPARENT LINEAR THERMAL EXPANSION COEFFICIENT AND AVERAGE APPARENT ELASTIC MODULUS OF INITIAL DESTRAINING FOR DIFFERENT PAPERS

Specimen	*Linear thermal expansion coefficient, α 10^{-5} per °C*	*Average initial destraining modulus,* Y kN/mm²*
Handsheet A	1·3	8·2
Handsheets B and D	1·8	7·3
Handsheet C	1·0	16·1
High-stretch sack paper, MD	1·8	7·3
Blotting paper	1·0	4·3
100 per cent rag paper, MD	0·6	16·8
100 per cent rag paper, MD, preloaded	0·6	15·6
100 per cent rag paper, CD	1·3	7·9
100 per cent rag paper, CD preloaded	1·3	6·9

* Arithmetic averages, the individual values vary up to ±15 per cent about the average value

Reasonable correspondence between the calculated thermal expansion coefficient and the reported values in the literature was also obtained for

other materials tested. Similarly, the approximate relationship between the calculated thermal expansion coefficient and the destraining modulus was observed, too, with cellulose film specimens and with the metal specimens.

The approximate inverse relationship between the thermal expansion coefficient and the extension modulus is a fundamental property of a wide variety of materials.[59] It can be explained by quantum mechanical considerations or by simple reasoning from the shape of the interatomic potential energy curve.

Residual change in internal energy after load-unload cycles

NOT all of the rate of work input during plastic region straining of paper or aluminium is changed into heat (Table 7). Fig. 10 describes the residual (net) change in the internal energy (see appendix 1) after load-unload cycles for the cross-direction (CD) specimens of 100 per cent rag paper. The work done during the straining and the hysteresis work—that is, the work lost in the load-unload cycle—are also illustrated. These three variables are presented as a function of the maximum elongation in the load-unload cycle. Previously unstretched specimens were used. The line segments give the 90 per cent confidence intervals for the average value of three to four specimens. (Stress relaxation took place before unloading.)

It is seen from Fig. 10 that the residual internal energy starts to deviate significantly from the zero value only after cycling to at least about 0·5 per cent. Similar results were observed for the machine-direction (MD) specimens and for the handsheets studied. About 30 per cent of the hysteresis work is stored as increased internal energy. In other words, about 70 per cent of the hysteresis work is lost as heat during the load-unload cycle. The latter value varied 70–80 per cent for the dry papers studied. The hysteresis work of 100 per cent rag paper CD is within 55–65 per cent, about the work of straining in cycles involving moderate to considerable amounts of plastic straining. This ratio varied 40–75 per cent for the various papers tested.

The respective results obtained with cellulose film indicated that there was practically no net heat phenomenon collected with a load-unload cycle. In other words, the residual change in internal energy was nearly equal to the hysteresis work, which was 50–65 per cent of the work of straining for cycles involving elongations of 2–4 per cent.

For aluminium foil, the hysteresis work was nearly equal to the work of straining and the residual change in internal energy was 30–40 per cent of the hysteresis work.*

* This figure may be only 10–15 per cent because of the possibility of an undetected heat flow in the case of aluminium foil

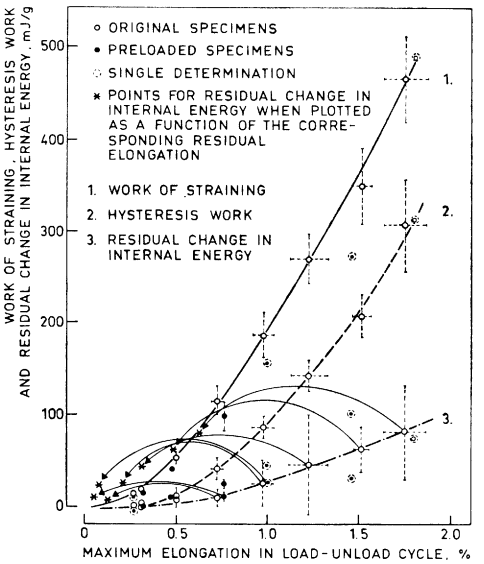


Fig. 10—Work of straining, hysteresis work and residual change in internal energy as functions of maximum elongation in load/unload cycle for 100 per cent rag paper CD

Role of residual stresses in change in internal energy

The change in internal energy after the load-unload cycle may involve stored strain energy besides structural changes. Because the specimen itself is in an equilibrium condition (no external load), the concept of stored strain energy means that some of the structural elements inside the specimen are under compressive stresses and some are under tensile stresses. For a cross-sectional element of the specimen, these stresses must be equal. There can be no net stored internal energy therefore due to the Kelvin thermoelastic behaviour.

The evidence for compressive stresses in the paper specimens after the load-unload cycle is mainly indirect. It is based on the premise that intrafibre secondary bond breakage (microrupturing) must be followed by some degree of rebounding. The following observations seem to support the existence of compressive stresses in the case of load-unload cycling of dry paper involved plastic deformation—

1. Fast reaching of the maximum rate of heat generation in the beginning of destraining (see Origin of thermal phenomenon).

2. Telescoping phenomenon—that is, the movement of cell wall lamellae and fibrils with respect to each other—
   (*a*) The stress relaxation of mechanically conditioned paper specimens is faster at low initial loads than that of original specimens.
   (*b*) If interfibrillar shear and breakage of intrafibre secondary bonds are accepted in straining of paper, it automatically means compressive stresses in destraining.
   (*c*) Evidence of some internal friction in mechanically conditioned specimens (see discussions of Fig. 4).
3. Stress recovery during a destraining pause.
4. Values of change in internal energy after cycling compared with the corresponding values of work of straining and the corresponding values of internal energy during straining indicated compressive stresses after cycling. The use of these comparisons is based on theoretical considerations concerning the role of residual stresses to the internal energy after the load-unload cycle. The respective results with cellulose film and aluminium were also used, together with literature information dealing with the role of residual stresses in these materials.

Based on this analysis, it is concluded that the paper specimens that have undergone 'apparent' plastic straining contain most probably a considerable amount of additional residual stresses compared with the original specimen.

Role of breakage of fibre-to-fibre bonds

SO FAR, the analysis of the results of thermodynamic behaviour of paper has not necessitated the use of fibre-to-fibre bonds as an important structural feature of paper. It could be said that fibre-to-fibre bonds have not made paper special in a thermodynamical sense.

The role of interfibre breakage was studied by measuring the change in the specific light absorption coefficient after straining to rupture in the microcalorimeter and by observing the corresponding behaviour of fibres in the rupture zone. The light scattering data are given in Table 6. The observed increases are in line with data reported in the literature. Thus, it is quite clear that even during plastic region straining of dry paper one generates some external fibre surface area, which scatters light. (It does not necessarily follow that these surfaces were in molecular contact previously.)

Incidentally, as was stated earlier, the residual change in internal energy starts to change from zero at about 0·5 per cent elongation in load-unload cycle. This is also the extension beyond which the straining causes a net change in the specific light scattering coefficient.[60]

One might now be tempted to conclude that the simultaneous occurrence of dissipative heat generation and irreversible increase in the specific light scattering coefficient proves that the plastic region of the load/elongation curve *is*

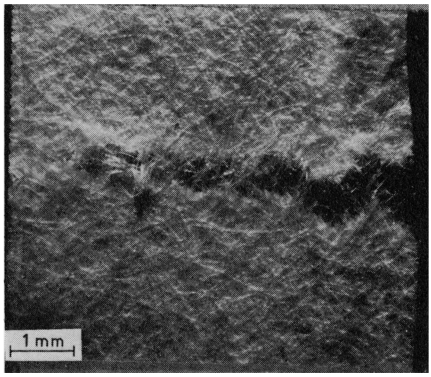
caused by the breakage of fibre-to-fibre bonds. This is not a proper conclusion, however, because it is not supported by other experimental results as follows—

1. The dissipative heat flow generation was similar for all papers—that is, independent of RBA (relative bonded area) or the length of the plastic region.
2. Generally, the heat flow just before the specimen ruptures did not show any unusual feature, even with the papers having low RBA.
3. Fibre-to-fibre bonds showed considerable residual strength in the final rupture and the behaviour of fibres and fibre-to-fibre bonds in the rupture zone was not directly related to the prerupture response of various papers.
4. Stress relaxation results indicated a similar mechanism for the whole stress range (see separate discussions). Besides, the initial and intermediate stress relaxation of paper was by no means special when compared, for instance, with cellulose film and aluminium.
5. The maximum of heat outflow was always reached faster in destraining than the maximum of the heat inflow during initial straining.
6. Some intrafibre bonds are as susceptible to local rupturing as are intrafibre bonds because of (*a*) similar geometry and similar physical binding forces, (*b*) the existence of misalignment zones, delaminations and lumen bonds in the cell wall and (*c*) the greater probability of molecular stress concentration inside the cell wall following the consolidation of the paper structure.
7. The initial destraining modulus after stress relaxation was higher than the initial straining modulus, especially if some apparently plastic deformation had occurred in the previous cycle.
8. Apparent permanent set was always observed after load-unload cycling involving 'apparent' plastic deformation. This observation ties in with the fact that, if the shape of the load/elongation curve during plastic region straining would be *mainly* due to the breakage of fibre-to-fibre bonds, then one would not see a pronounced permanent set after unloading the specimen.[61]
9. Paper and aluminium had very similar general thermodynamic behaviour (mechanical and thermal). It is well known that the irreversible phenomenon in aluminium is caused by slip between the structural elements. There are plenty of possibilities for some kind of slip to occur inside the cell wall (see point *6*).

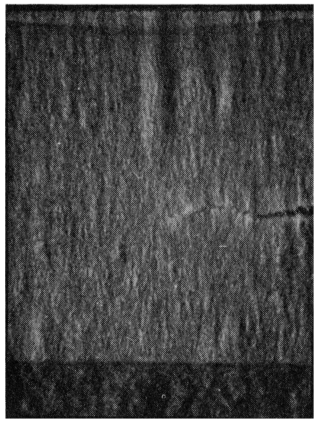
Based on the above reasoning, it is concluded that the plastic region in the load/elongation curve of paper *is not caused* by the breakage of fibre-to-fibre bonds, but it is connected with the significant irreversible intrafibre deformation at the molecular and supramolecular level of the cell wall structure. At the same time, some interfibre bond breakage does occur, though it is a natural extension of the above-mentioned intrafibre plastic deformation. Page & Tydeman earlier arrived at a similar conclusion.[29]

Partial breakage of fibre-to-fibre bonds

In order to put the interfibre and intrafibre deformation into a right perspective with each other, attention is drawn to Fig. 11. This picture describes the opacification observed in straining the highly refined free-dried handsheet B_m (Table 6). The rupture elongation of this sheet was 14 per cent when tested at 50 per cent rh and the shrinkage during drying was 12 per cent. The increase



ENLARGEMENT OF A TYPICAL RUPTURE LINE



UNSTRAINED AREA BEHIND THE CONTACT LINE OF THE CLAMP

Fig. 11—Partially ruptured tensile specimen of handsheet B_m

in the scattering coefficient was 130 cm^2/g. The ultimate rupture was so slow that one could stop the straining before a complete separation had occurred. (This means that there was very little strain energy stored in the specimen at the moment of final breakage; the specimen had undergone an extensive plastic deformation.)

It is clear that the drying shrinkage had to be 'pulled out' before the rupture elongation was reached. This result is similar to that reported by Helle.[30] It is also well in line with the concept of Page & Tydeman,[29] who proposed that the deformation of paper can be visualised to some extent as the reversal of the shrinkage process—that is, removal of the microcompressions from the bond sites and between them.

The creation of light-scattering surface was so great in the case of handsheet B_m that the difference in the opacities of the unstrained and ruptured specimens could easily be seen (Fig. 11). The unstrained area behind the line clamp is seen to be somewhat translucent, whereas the area strained to rupture is opaque. The immediate area surrounding the partial rupture line and the tip of this line is even more opaque than the rest of the strained area. It is obvious that there must have been some breakage of interfibre bonds during this straining.

Yet, when one observes the fibres in the rupture zone, there are not very many fibres that would have been pulled out intact. Most of the fibres have broken with short free segments, indicating strong residual bonding between the fibres during the rupture. Fig. 11 and similar results with other paper specimens led to the conclusion that the bond breakage *is mostly partial.* The obtained result agrees with the direct observations made by Page and coworkers.[44]

The results obtained are actually quite natural. Especially in a freely dried sheet, the fibres wrap around each other under the action of the surface tension forces and by the shrinkage action of the web caused by the transverse shrinkage of the cell wall at the bond sites. When the sheet is strained, the forces transmitted from one bond site to another will first load the part of the interfibre bond that was formed last, thus the straining forces will try to unwrap the free segments. Furthermore, one would expect that the periphery of typical interfibre bond to be weak. This is because it was formed at a later stage during consolidation and because it does not involve such molecular and elementary fibril entanglement as the original contacts between swollen cell walls, which were formed in the beginning of consolidation. The forces acting will try to peel the fibres loose and will simultaneously produce stress intensifications at the periphery of such interfibre bonds, where considerable wrapping of fibres is involved. Thus, the peripheries of these interfibre bonds are probably quite susceptible to rupture.

Note on Nordman's bonding strength values

NBS value is calculated by dividing the work lost in a load-unload cycle (the irreversible work) by the corresponding increase in the external area of the fibres of the specimen. In other words, NBS $= \Delta W^h/\Delta S$, where ΔS is the increase in surface area (assuming that the light scattering results are transferred to molecular surface area by a suitable proportionality factor).

If a considerable part of the work lost in a load-unload cycle is changed into heat, however, it is not proper to use the work lost in cycling in computing the NBS value. Instead, one should use the net change in internal energy in those computations. This means that the new NBS values are only 20–30 per cent of the old ones. Yet, because a large amount of the net change in internal energy may be due to residual stresses, even the use of net change in internal energy is questionable.

Let the net change in internal energy be used to estimate the NBS value. Let also a proportionality factor of 100* be used between the increase in the gas sorption surface area and the increase in the specific light scattering coefficient. Then one finds that the respective NBS values for handsheets A, B and C (Table 6) are 460, 850 and 800 erg/cm^2. The NBS values obtained are still about an order of magnitude greater than the measured or predicted free surface energy values of cellulose materials.[63, 66] The difference can very well be due to the stored strain energy.

Interpretation of results in terms of paper structure

DESPITE wide variations in the structure of the sheet, all papers made from natural cellulose fibres showed a similarity in the form of the curve for heat flow during straining. This similarity may be called fundamental: it implies that all papers have similar deformation mechanisms.

The initial straightline response is governed by energy elastic deformation. No significant frictional force component is present, neither is there a significant entropy elastic force component present. These conclusions are derived from the reversibility and sign of the heat phenomenon. The latter conclusion is quite natural if one considers the relatively tight packing of cellulose chains in the crystalline and amorphous regions of the cell wall.

The observed results of the increase of the destraining modulus and the respective decrease in the thermal expansion coefficient between freely and restraint-dried handsheets (B and C, Table 8) clearly show the importance of stress equalisation in the amorphous region of the cell wall. It is felt that the hypothesis attributing the increase in the modulus of tension-dried paper

* Here, one has used Barber's[62] result that the new BET surface area is about three times the old BET surface area with a proportionality factor of 33 between the old BET area and the change in scattering coefficient[51]

mainly to (*a*) orientation of fibres, (*b*) to better equalisation of stresses between the fibres and (*c*) to fibril alignment underestimates the role of simultaneous molecular and supramolecular structural changes inside the cell wall. In other words, the tension drying makes the matrix material less amorphous. Because mechanical conditioning did not change the estimated thermal expansion coefficient, it is concluded that the structural changes due to plastic straining are insignificant compared with the structural changes during consolidation.

The dissipative heat generation during the straining of paper in the plastic region proves that irreversible structural changes are occurring during this straining. Thermodynamic analysis cannot predict if these structural changes are *mainly interfibre* or *intrafibre* phenomena or if both changes are involved. As has already been discussed, critical analysis of experimental data and previously published results of the deformation mechanism with other materials has led to the conclusion that significant irreversible intrafibre deformation controls the plastic region behaviour of paper. Although it is impossible to quantify how much of the energy consumed during straining is used in *intrafibre* processes, it appears that, except for the magnitude of the change in the light scattering coefficient, the observed results for dry paper could be accounted for to a large extent by irreversible intrafibre deformation.

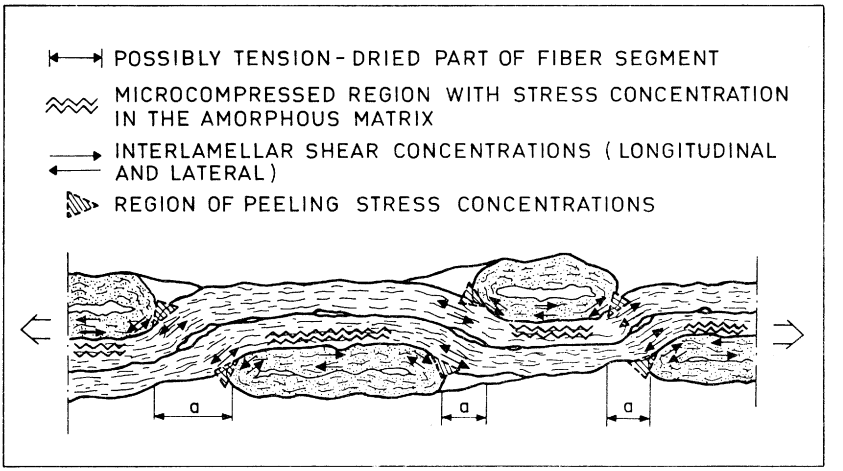


Fig. 12—Schematic representation of some possible stress concentration regions in apparent plastic straining of paper

The dissipation of work takes place when molecular stress concentrations cause the secondary bonds of the cell wall structure to yield. These stress concentrations are in many cases related to interfibrillar shear deformations. Fig. 12 shows schematically some possible regions of stress concentrations.

The appearance of these stress concentrations is affected by various structural features at all levels of the structure of paper (Table 1). The breakage of interfibre bonds has an important bearing on the stress redistribution, because relatively large structural units are involved in comparison to intrafibre bond breakage. Besides, interfibre bond breakage does not lead to a bond reformation as is the case most probably with intrafibre bond breakages. Thus, a breakage of fibre-to-fibre bond (partial or complete) leads to an intense dissipation of work in a new position not necessarily connected with a region of previous intense dissipation. Simultaneously, fibre segments that were previously less active are most likely to engage in sharing a greater part of the load.

The proposed hypothesis resembles closely the matrix model,[32, 35] which itself is based on the fundamental role of microcompressions in consolidation and the subsequent behaviour of paper.[29]

The results obtained have some important implications on papermaking. Throughout various phases of papermaking, structural heterogeneities are introduced into the cell wall of individual fibres. These 'flaws' increase the probability of serious stress concentrations during the straining of paper and thus lower the region of elastic response. Design of processes that create fewer flaws and of methods for correcting the effect of flaws ought to obtain greater attention in papermaking research. One the other hand, in some applications of paper, the existence of cell wall flaws means that the heterogeneous structure of paper (formation plus summerwood or springwood) can dissipate energy and it can be extended without rupturing.

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***Appendix 1**—General thermodynamic aspects of deformation*

THE deformation of any material requires energy. This energy is usually supplied in the form of external work. According to the first law of thermodynamics—

$$\Delta E = \Delta W\uparrow - \Delta Q\downarrow \quad . \quad . \quad . \quad . \quad (1)$$

where ΔE = change in internal energy
$\Delta W\uparrow$ = (net) work done on the system (= specimen)
$\Delta Q\downarrow$ = (net) heat given out by the system (= specimen)

It is possible to separate four classes of extension based upon the relationship between the work of extension and the accompanying heat of extension (Table 9). In the first class, the internal energy of the specimen increases during extension both from the work and heat contribution. In the second class, the internal energy is still increasing due to extension.

In the third class, one has three possibilities—(*a*) The deformation is totally irreversible; all the work done is dissipated as heat; no recovery is possible; (*b*) The internal energy varies throughout the material in such a manner that the average internal energy remains constant; (*c*) The internal energy does not change, not even locally and the work is done against the thermal kinetic forces (entropy force). In the two last cases, the material possesses a potential for the recovery of the deformation.

TABLE 9—VARIOUS PHENOMENA OF THERMODYNAMIC BEHAVIOUR DURING STRAINING

1. $\Delta W\uparrow > 0$	$\Delta Q\downarrow < 0$
2. $\Delta W\uparrow > 0$	$\Delta Q\downarrow > 0$, but $< W\uparrow$
3. $\Delta W\uparrow > 0$	$\Delta Q\downarrow = \Delta W\uparrow$
4. $\Delta W\uparrow > 0$	$\Delta Q\downarrow > W\uparrow$

In the fourth class, the internal energy of the specimen decreases during straining. This could happen, for instance, during crystallisation or phase change induced by straining.

If one carries out a cyclic deformation (straining and destraining), it is possible to separate three kinds of thermodynamic behaviour (Table 10)—(*a*) elastic behaviour (that is, mechanically and thermodynamically reversible); in which case, the net heat given out and the net work done are zero, thus there is no change in the internal energy; (*b*) anelastic behaviour (that is, mechanically reversible and thermodynamically irreversible), in which case, the net work done is positive, the net heat given out is positive and their numerical values are equal, thus all the irrecoverable work is dissipated as heat that flows out of the specimen and there is no change in internal energy due to cyclic deformation; (*c*) mechanically and thermodynamically irreversible. In this latter case, the change in internal energy caused by cyclic deformation is not zero and there is a net work lost—that is, an irrecoverable work and a net heat given out—but these are not equal in magnitude.

Examples of these three classes are—(*a*) Hookean behaviour, (*b*) cyclic straining of a specimen showing hysteresis (internal friction) and (*c*) straining leading to permanent set (plastic deformation) (Fig. 13).

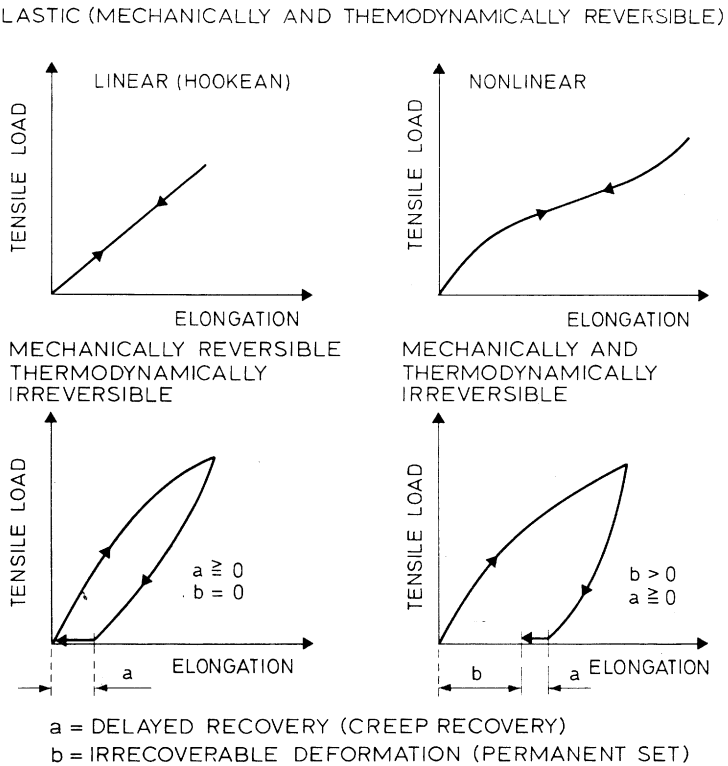

Fig. 13—Different phenomena in uniaxial extension

TABLE 10—THERMODYNAMIC PHENOMENA ASSOCIATED WITH CYCLIC DEFORMATION

1. Elastic	Mechanically and thermodynamically reversible	$\Delta E = 0;\ \Delta W\uparrow = 0;\ \Delta Q\downarrow = 0$
2. *Anelastic*	Mechanically reversible thermodynamically irreversible	$\Delta E = 0;\ \Delta W\uparrow > 0;\ \Delta Q\downarrow = \Delta W\uparrow$
3. Plastic	Mechanically and thermodynamically irreversible	$\Delta E \neq 0;\ \Delta W\uparrow > 0;\ \Delta Q\downarrow \neq \Delta W\uparrow;$ $\Delta Q\downarrow > 0$

Appendix 2—*Kelvin's thermoelastic equation*

ONE of the Maxwell relationships of classical thermodynamics states—

$$\left(\frac{\partial S}{\partial P}\right)_T = -\left(\frac{\partial V}{\partial T}\right)_P \quad . \quad . \quad . \quad . \quad (2)$$

Using the definition of entropy for a reversible process, one obtains—

$$\delta Q = TdS = -T\left(\frac{\partial V}{\partial T}\right)_P dP \quad . \quad . \quad . \quad . \quad (3)$$

Further, for uniaxial isothermal deformation, one obtains—

$$\delta Q = T\alpha l \delta f \quad . \quad . \quad . \quad . \quad (4)$$

where the identities or approximations $\delta V = A\delta l$; $A\delta P = -\delta f$ have been used and $(\delta l/\delta T)f \cong (\partial l/\partial T) = \alpha l$. It is also assumed that Poisson's ratio is not close to 0·5.

Equation (*4*) is one form of Kelvin's thermoelastic equation. It states that, in an isothermal uniaxial extension, a specimen having $\alpha > 0$ receives heat from its surroundings and releases heat to the surroundings during the removal of the force or during a compressive deformation.

Using the first law of thermodynamics, equation (*1*), one has for a unit volume of specimen following Hookean and Kelvinian behaviour in straining—

$$\Delta e = \Delta w \uparrow + \Delta q \uparrow = 1/2\, Y\varepsilon^2 + T\alpha Y\varepsilon \quad . \quad . \quad . \quad (5)$$

and

$$\Delta \dot{e} = Y\varepsilon\dot{\varepsilon} + T\alpha Y\dot{\varepsilon} \quad . \quad . \quad . \quad . \quad . \quad (6)$$

Equation (*6*) states that, during the constant rate elongation (initial straightline response), the internal energy is increasing from two terms—(*a*) the thermal contribution, which is independent of elongation (that is, constant) and (*b*) the elastic energy contribution, which is a linear function of strain. In the beginning of straining ($\varepsilon < T\alpha$), the thermal contribution outweighs the elastic (mechanical) contribution.

Kelvin's thermoelastic equation, equation (*4*), allows one to obtain an estimate for the linear thermal expansion coefficient of the tested material. In other words, by plotting pairs of δQ and δf of an elastic deformation, one should obtain a straight line with a slope equal to $T\alpha l$. In this work, data from destraining has been used to estimate the linear thermal expansion of various materials.

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Discussion

Dr M. B. Lyne Dr Page mentioned in his contribution some work that we had carried out previously (not the subject of this paper) on the relationship of in-plane and Elmendorf tearing strength to printing press runnability.[1]

First of all, we made a high speed ciné study of the problem areas in a large letterpress works in Vancouver. We observed that the trouble spots were associated with uneven stress or stress concentration—angle bars, the slitter, the folder and the reel stand when rolls with a tight edge were being run. In the latter case, the press operators could only go so far in cocking the roll in order to make it run true before significantly increasing the probability of pressroom breaks.

I would also like to offer a simple illustration that may throw some light on the nature of the pressroom break. If you consider a 1 in wide strip of newsprint, the force required to rupture it in tension is approximately 100 times the force required to propagate an in-plane tear across the paper (with our tearing geometry). Considering a wide newsprint web in a printing press, the force required to rupture it in straight tension would thus very much exceed the force required to propagate a tear across the web. This convinces me that printing press breaks are essentially tear phenomena, since tear-initiating defects such as edge and shive cuts are always present.

In comparing the Elmendorf and the in-plane modes of tear to a printing press break, we made the comment that the printing press cannot possibly rupture the sheet of paper as is done in the Elmendorf case. If you examine the stress applied to the web in the printing press, you will find it is impossible to have stress applied in the Elmendorf fashion. The Elmendorf test is involved with very serious bending forces and characteristic delamination. Delamination, too, does not occur in printing press breaks.

The Chairman May I ask if you have actual experience that the in-plane test gives better correlation with press room performance than the Elmendorf test does?

Dr Lyne We were seriously interested in this, of course, since it is the

heart of the matter. In response to that interest, we created the pendulum in-plane tear tester. The original Instron instrument that Dr Van den Akker and his co-workers designed and that formed the basis of our initial studies was too slow to be used for a large-scale study of newsprint. To establish the relationship between a test and runnability needs having the test used on an enormous number of rolls, then to trace their runnability in the press room. We are in a position to do that now. The pendulum instrument should allow normal mill testing on a large number of rolls. I might add that the prototype of this instrument has actually been tested in a mill test station and it was found that it could be operated with the same ease and speed as the conventional Elmendorf tester.

Dr V. C. Setterholm Some years ago, we observed that at equal levels of restraint during drying elongation at rupture in paper appeared to be independent of fibre orientation. I have never had an explanation for it until today, yet I think it to be consistent with the behaviour of paper that Dr Ebeling has described.

Dr K. Ebeling I am sorry not to have included the failure mechanism in my study. Some data in my thesis work has not yet been fully analysed. I personally believe that there is not very much connection between the failure mechanism and the prerupture behaviour of paper.

The situation you indicated could arise with highly refined pulp fibres and from drying restraints involving a moderate to considerable amount of wet stretching before drying commences. I believe that, with slightly or moderately beaten fibres and no wet stretching, the anisotropic shrinkage potential of the cell wall and the anisotropic elastic properties makes the elongation at break depend on fibre orientation.

Dr D. H. Page I want to reply to the earlier comments of Lyne and Ranger on the skew tensions in the press. If you have a sheet running between rolls, you really have little opportunity for angular distortion. You might have a lot of skewness with respect to tension (perhaps one edge is slack and the other highly strained), but the angular distortion would be only about half a degree. The fracture resistance as we measure it would be quite appropriate. The angular distortion in the in-plane tearing test is 12° and this is as unrealistic in usage as is the Elmendorf tearing test.

Now, with strain rate, I think there is a misunderstanding of the term quasi-static fracture energy. It is so called, but that does not mean to say that it happens slowly, merely that there has to be insufficient energy stored in the jaws and the specimen for completion of fracture, so that the energy comes

from the applied strain. The straining can be completed as quickly as you like. The specimen length has to be short to satisfy these conditions anyway, so it is simple to make the strain rate fairly high. We are studying the effect of strain rate.

Dr Lyne I agree completely with the analysis Dr Page has made for paper between parallel rollers in a printing press. I believe this corresponds to our edge tear test. As I mentioned, however, the problem areas in the press seem to be such spots as angle bars, the folder and the slitter. In these cases, you can indeed talk about angle (not necessarily 12°) if you consider the zone around a web defect such as a cut.

Dr S. I. Cavlin I would like to draw your attention to the fundamental difference between fracture energy testing and the in-plane tear testing, which I think is relevant for the runnability and its characterisation.

In a quasi-static fracture process, the energy needed to create a new surface is continuously added to the strained material during the surface creation. In a brittle fracture process (which Griffin considered), the fracture energy comes from the strained material itself. If the elastic energy stored in the paper web is not enough, when the failure is induced, just part of the web will be fractured in a brittle mode and the rest in some mixed brittle/quasi-static mode. The stored elastic energy per unit cross-sectional area is proportional to the span length and the stress. Thus, for every failure stress, there is a critical span length for brittle fracture. This span length must be important for runnability. At normal breaking stresses, the critical length is about 50 mm; at smaller stresses, in the order of 1–5 m. The conclusion is that the crack propagation energy is important, but also the span length and the loading. In the in-plane tearing method, these variables are mixed in a ratio that is unknown. It is of course best to separate the effects.

Mr J. R. Parker There are two questions I would like to ask of a practical nature. Firstly, I recall some (unpublished) work I did several years ago on the effect of shives and long fibres in handsheets. I divided some pulp into two halves, from one I removed the long fibres and shives. Handsheets were made, some were calendered and the reduction in bursting strength measured. There was very little reduction for the sheet without the shives. I wonder if shives could account for Dr Moffatt's observations.

The second point arises from an experiment done to find out whether tearing strength was of any relevance in preventing the spreading of a small crack in paper. I took paper samples about 150 mm long, 90 mm wide, of a variety of paper grades ranging from newsprint to greaseproof cut in both

machine and cross directions. Their tensile strength was determined, with and without a transverse 10 mm slit in the centre. A plot of the slit against the non-slit tensile strength gave a straight line. Now, it is noteworthy that the points lay on the line even though the Elmendorf tearing strengths across the samples varied enormously. There seemed to be no tendency for the plotted points to vary in any systematic way in relation to tearing strength.

Dr Lyne I would point out that Elmendorf tear does not relate to the kind of test you made. In the Elmendorf test, several sheets are torn at once and the tear line follows some sort of mean path rather than propagating freely. Of course, the application of stresses are also quite different in the Elmendorf tear and in your test.

Dr J. M. Moffatt Firstly, the results of tensile strength with slit and non-slit confirm what I presented on Tuesday.

Secondly, I agree that shives are very important, not just large shives, but even small shives, causing local weakening. Another point to emphasise is that a sheet and the shives are not two separate and distinct elements. Shives are merely part of the distribution of fibre sizes and must be considered as an integral part of the pulp mix from which you start. Take the shives out to change your pulp and you change your starting material, for the removal of shives is what you should be doing as far as possible. Unfortunately, we seem to have arrived very close to the point of diminishing returns in this direction. I will have to think about the significance of the tear.

Mr U. Ullman We have collected some information from handsheets made out of various pulps and pulp mixtures ranging from 100 per cent groundwood to all-chemical pulp with different degrees of beating. For papers with low chemical pulp content and a consequent low extensibility, the breaking loads are strictly proportional to the width of the load-bearing part of the sample and thus fall along the curve Mr Parker has shown. When, however, you test more extensible papers (for example, with high chemical pulp content), the breaking loads will fall within the high range of Mr Parker's graph. Because of the extensibility of these papers, stress concentrations will occur around the ends of the slit.

This will cause the slit samples to break at loads lower than those proportional to the load-bearing width of the samples. The line in Mr Parker's graph will therefore, at higher breaking loads, bend towards the axis for breaking loads of unslit samples.

All pulps and pulp mixtures followed this schema falling on one and the same curve. Thus, we (like Mr Parker) have not been able to identify an effect

that can be referred to as flaw-carrying ability. A similar effect can occur, however, because of difficulties in forming the slit. Papers with a low chemical pulp content form brittle webs. In such webs, the slitting operation frequently gives micro-cracks protruding from the ends of the slit, unless extreme care is exercised. Such micro-cracks in weak samples might be the cause of effects that can have been interpreted as flaw-carrying ability.

Mr Parker It is some time since I did this work, but I am fairly certain we used not just newsprint, but greaseproof too, which extends considerably. Yet the points all seem to lie on the line and there was no systematic change with tear. I am sorry, I did not observe the effects Mr Ullman expected.

Dr D. Atack We seem to accept arbitrarily defined rankings of runnability without much question. For instance, about ten years ago, the runnability of newsprint was almost exclusively identified with the groundwood shive content and the type it contained. More recently, Steinberg of the *New York Times* noted that in his experience only a very small percentage of press room breaks could be attributed to the presence of groundwood shive and that many press room breaks were due to other factors such as defects introduced during handling. He suggested that the more precise control of moisture content across the reel may be much more important in leading to better press room runnability.

I mention this example to indicate two extreme views about the possible effect of one particular source of poor runnability in one particular grade of printing paper. I believe it is quite difficult to quantify the contribution of individual paper properties to performance on the presses. In general, I suggest that clearer definitions of end use defects are needed before they are related, willy-nilly, to somewhat more clearly defined and idealised fundamental paper properties.

Prof. J. Silvy In a normal sheet of paper submitted to a tensile load, we have stress concentrations and their amplitude can be very high, maybe 4–5 times the mean stress concentration distributed over the section of the sheet. Due to these 'natural' stress concentrations, the value of the breaking load of a strip of paper can be the same whether or not (as Mr Parker did) you initiate small cracks in the strip. Where there are cracks in the sheet, you find the same results as without cracks, which means that 'natural' paper breaks because of stress concentrations.

The importance of the stress concentrations in the rupture of paper have been discussed previously (for instance, *Paper Tech.*, 1971, **12** (5 & 6), 390–392).

A second point is about tear and I would like to report some results observed. If you take a sample of paper and break it by strain, then take the pieces of the sample and measure the out-of-plane tearing strength, you will find that the tearing strength is exactly the same up to the rupture as if the paper had not been loaded before. What is the energy consumption of the tear? By loading these samples in the first experiment, we have spent much more of the potential consumption energy of the paper; this energy seems to have nothing to do with the energy consumed during the tear.

Dr Page I want to return to the comments that were made the other day on fracture resistance and the in-plane tearing test. The question was left why 12° was chosen for in-plane tearing test. Perhaps Dr Van den Akker could answer that in a few moments, but my recollection was that, if you make the in-plane tear angle 12°, then the result is insensitive to angle; it makes no matter whether it is 11° or 13°, you will still get the same value. That may be so, but it does not mean to say that we are measuring some kind of fundamental property when we do that.

I would like to say what happens in these two tests of fracture. In each case, there is a stress concentration in the form of a circle at the tip of the crack and, over that region, the yield point of the paper is exceeded. Plastic work is being done. The tip advances and further plastic work is done. That is where the energy goes in those tests.

In the in-plane tearing test, the stress concentration is higher than in the fracture resistance test, so that only a small region is yielded and the energy consumed is lower. If you plot in-plane tear values against angle from 0° to 12°, you find that the energy falls off very rapidly over the first few degrees and reaches a steady value at 12°.

There is, however, no relation between the in-plane tear value at 0° (which we call fracture resistance) and the in-plane tear value at 12°.

I have one other point on runnability. I am not sure whether we are discussing this or the basic properties of paper, but fracture resistance is certainly a basic property of paper. It is the energy required to propagate a crack under plane stress. Whether or not it is correlated to runnability, of course, is another question. It is probably correlated to some usage properties of paper. If you have a small nick in a paper bag, the very first propagation of that crack is probably in the plane stress opening made, so that fracture resistance measured in this way may be a good criterion of the failure. Obviously, there are many other factors that might affect runnability in press rooms.

Chairman My conclusion from the meeting here has been that it is

perhaps not the strength of the paper itself that is important for runnability, but the number of defects. This, of course, is an old thinking. There are always stress shocks in the pressroom; if there are defects in the paper and they occur at the same time as the shocks, there will be a break. These defects could be the result of the heterogeneities in the grammage, it could be edge defects or the presence of shives.

I want to add one more kind of defect. When I say that perhaps it is not the strength itself that is important, then immediately the comment is made that papers with semi-bleached kraft pulp are stronger than that made with sulphite pulp. Obviously, the strength of the paper is relevant, but it could be in the following way.

We have to remember that a sheet of newsprint is a very thin sheet, considering only the fibres forming the network proper. We have to remember that there are very few real fibres in mechanical pulp, because half of the pulp is fine material. These fibres have been obtained in a mechanical yield of 100 per cent. Therefore, the number of fibres in 1 g pulp (the fibres fraction) is only half of that in the corresponding amount of a chemical pulp. The number of fibres in a sheet of newsprint with 15 per cent chemical pulp, therefore, corresponds to a sheet made from ordinary chemical pulp of only 20 g/m^2, so it is a very thin fine network. When we run this paper through the open draws of the papermachine, the network will expand to some extent and defects can be introduced into the sheet. The kind of chemical pulp will definitely influence the wet strength of this web.

Dr J. A. Van den Akker Referring to Derek Page's discussion of the difference between in-plane tear and his fracture resistance energy, I think I disagree. Basically, we are dealing with the same phenomenon in both of these tests. In my opinion, we do not have an appreciably different area of dissipation of energy from that existing in your test.

There is another factor that enters the picture. Part of the scientific method is that one tries to separate the variables, so the more fundamental of these two tests is the in-plane tear with an angle of, let us say, 12°.

If the angle is essentially zero, more tensile load must be applied to the sheet. This means that more energy is irrecoverably dissipated in the sheet, thus the observed energy increases with decreasing angle.

From a practical point of view, this steep slope in the energy graph plotted against angle curve (at small angles) is undesirable. Although it is possible to make an instrument in which we need not worry about the jaws deviating from parallelism, the non-uniformity of paper will in my view have an effect equivalent to variability of angle and that, accordingly, there might be undue variance in the test result. I emphasise that my main point against the zero

angle is the undue irrecoverable energy dissipated in the sheet because of the appreciably higher tensile loading that is required.

Prof. Silvy's question is one often logically raised. Once an ordinary tensile break begins, the failure occurs extremely rapidly and the energy for the tearing is not sensed by the instrument. The instrument cannot be fast enough to measure this; the energy for the rapid tearing comes from the strain energy stored in the strip. (If the span length is reduced to a very small value, as in our work published in 1958 on the importance of fibre strength, then there is not enough strain energy stored in the strip to continue the break.)

Dr C. J. Edwards As a member of the printing industry, one of the main users of paper, I have been a little disappointed so far that work in the area of the mechanical properties of paper has tended to avoid impulse studies in which stresses and strains are applied at very high rates. This is very important, since I have heard it said among machinery manufacturers that paper seems to be able in a printing press to go beyond the stresses under which one would normally expect it to break on the evidence from static tensile tests. Perhaps this is the experience of many people.

Another very interesting field to study is when paper is subjected to heat rapidly. We examined this briefly about three years ago, as it is very common now in the printing industry, with inkdrying having to take place at rapid rates. Heat is normally applied by infra-red irradiation, by gas flames or by high velocity hot air, the surface temperature of the paper reaching perhaps 200°C. We found by studying gas flames and infra-red that, certainly within less than 2 s, the cross-direction of the paper shrunk by up to 0·5 per cent and the moisture content of that paper had gone down below 1 per cent, probably even down to 0·5 per cent.

We also found that, if moisture was subsequently added to the paper rapidly, the paper did not recover its original dimensions and the amount of recovery seemed to depend on the rate at which the moisture was added.

Unfortunately, other priorities prevented a detailed study and I believe that it would in any case be better studied in a research institute or perhaps at a university.