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IMPACT OF FUNDAMENTAL RESEARCH ON KNOWLEDGE OF THE MECHANICAL PROPERTIES OF PAPER

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Synopsis

Recent trends in structural design theory and material science are reviewed and discussed in relation to research on the mechanical properties of paper and board.

The purpose of materials science is, apart from general scientific curiosity about the structure and behaviour of matter, to form a basis permitting the solution of structural design problems.

Applied to the man-made engineering material paper, we are thus interested in scientific methods and results which permit, for instance, the design of flexible or rigid paper packages. Our interest includes also the mechanical behaviour of stationary or moving paper webs both on the paper machine and in various paper processing techniques and fields of use.

Our ultimate goal is twofold. On the one hand we want to be able to predict the behaviour of the paper under various conditions from a (preferably) small set of mechanical properties. On the other hand we need a scientific basis to guide us in choosing raw materials, setting parameters in the paper making process etc., in order to design a paper fulfilling certain mechanical specifications.

It is important to realise that development in the general structural design field has only been possible by theoretical approaches combined with a large input of engineering intuition, aided by a vast background of practical experience. Key

experience data are laid down in various engineering codes. Studies of models and prototypes have always been needed to defend and justify new ideas and solutions.

If, for the moment, we neglect the very essential contribution from engineering intuition, we can say that structural strength problems and practical progress in understanding materials have come about by bracketing theoretical approaches on one hand with experimental work on the other.

Until quite recently the classical laws of elasticity applied via the laws of classical mechanics were the basis of most theoretical approaches. The application was to a very large extent one- or two-dimensional, with some displacement constraints in the `thickness´ dimension.

This method has been applied to increasingly complex structures like laminates and various composite materials. The classical methods had considerable difficulty in dealing with time-dependent phenomena. While many elastic materials can reasonably be treated with the ideal Hookean law, Newton´s viscosity law is hardly ever applicable to time dependent phenomena in condensed matter.

The advent of digital computers with increasingly higher speed and memory capacity has changed this situation. Through various discretisation methods based on matrix algebra and mesh formulations by both classical and modified Eulerian and Lagrangian methods, it is today possible to handle not only consecutive equations of spatial discretisations but also time integration. This will permit inclusion of time-dependent phenomena, like plasticity and visco-elasticity. This implies that the classical Hookean theory is replaced by a set of consecutive equations relating stress (pressure), to strain (volume change), strain rate, and also strain history. Eventually this method will include consideration of deteriorating structures and the behaviour of defects. It will then be possible to deal also with fracture mechanics.

Conventional structural analysis has some of its main problems when dealing with impact. Most classical dynamic theories are limited to cases of vibrations.

Some impact case analysis using FEM (Finite Element Methods) and FDM (Finite Difference Methods) has had practical significance, especially when there has been no possibility of carrying out experiments. Typical examples are found in safety management (e.g. simulations of the human body in accidents, or of a big plane crash into an atomic reactor).

The very important field of crack propagation is another field of active research. Crack growth is characterised by intense plastic deformation in very small regions. One approach is to assume that there exists one parameter which uniquely determines the state of stress and strain outside this zone and which can be related to external boundary conditions, essentially geometry and load. Other theories are purely probabilistic. Considerable computer work is done on models representing various ductile materials.

The application of all methods is severely limited by lack of experimental data. The problem is that there is at the moment no general agreement on which set of differential equations is most suitable to work with. This results in the dilemma that the experimenter does not know which material-specific parameter values are required.

One thing is clear. The classical set of parameters, based on elasticity theories, no longer have the same interest, because frequently the differential equations used do not involve Hookean behaviour.

One can expect a considerable period of flux until a new set of principles will be agreed on. The world will for some time again be as untidy as it was before the theory of elasticity became an accepted basis for analysis.

Eventually results of value will emerge even for materials like paper, which is far from Hookean and where the properties vary with temperature and humidity[9].

At this instance it is worth while to consider some results of fundamental research in the general science of materials. Such studies have only to a limited extent been confined to studies of mechanical properties. The behaviour of matter is more easily analysed by optical, electric and magnetic methods

than by direct mechanical methods. Mechanical properties are often deduced from theoretical relations with other properties.

The most important results in the last decades have come from studies of crystals and glasses. It has become increasingly evident that idealised structures, like structureless glasses and perfect crystals, are of less interest than earlier assumed. Most properties, including the mechanical, are dramatically influenced by minute imperfections, caused either by the presence of "impurities" or by structural defects on the atomic scale. The development of solid state physics rests on such knowledge. Studies of disorder in what is called ill-condensed matter, is one of the most active research fields in physics and materials science. Its advance can be traced to ideas from Andersson, Mott, Hammersley, and Broadbent, and terms like percolation and localisation permeate recent literature. The development of Markov processes and ergodic theory, probably the most active sectors in mathematics, has been of great importance.

Until now this development has had relatively little impact on the field of polymer materials. Most of the work is still based on theories of Flory and ideal rubber elasticity models. The percolation concept has however, recently been applied to typical polymer mechanical properties like vulcanisation and gelation. The French school of polymer physics working with new field concepts (scaling laws) has until recently been isolated because their methods have been strange to most polymer scientists.

The perplexing property of condensed matter, the bonding of atoms and molecules, has undergone several scientific revolutions in the Kuhn paradigm sense.

The first was the quantum mechanics revolution. This has been highly successful in interpreting the bonding of the atoms in chemical compounds. It has also contributed to the understanding of inter-molecular phenomena. The hydrogen bond concept is a typical example.

It suits the paradigm of a chemist to think of the behaviour of condensed matter as being caused by a relatively simple system of various kinds of localised bonds. If the chemist is working

on paper properties he will think in terms of the paper bond.

This is not the only possible model. Let us for example consider the forces which resist the separation of fibres surrounded by more than a molecule of water (initial wet strength problem). It is natural for a chemist to assume that a force is transmitted between adjacent fibre surfaces through chains of bonds including the water molecules. A typical exmple of this approach is depicted by Caulfield[10] with the bonds drawn as helical springs.

The existence of a structure which can transmit forces can only be deduced from the behaviour of the system when it is deformed.

A structure is then defined as the existence of a train or cluster of particles extending through the structure when it transmits forces. We cannot preclude the possibility that the structure does not exist when no forces are applied. If under straining the cluster does not extend throughout the system we are dealing with an assembly of independent particles or small clusters. It is not necessary that all distinguishable elements are members of the infinite cluster. It is not even necessary that any particle is a member of a cluster all the time, only that the infinite cluster exists at all times. If the strain increases, eventually the cluster will break and with it the structure.

Let us apply this to a suspension of large hard spheres. If the particles are small and exhibit Brownian motion there is hardly ever any contact between the spheres. But if the spheres are large and in a shear field two particles may make contact and remain in contact for a finite time. At a certain critical concentration, a finite fraction of the spheres will belong to an infinite cluster of spheres in contact. Cox and Mason[11] have demonstrated the surprising fact that hard macroscopic spheres above a critical concentration exhibit plug flow. In this regime the particles, although spherical, form an anisotropic structure. Similar plug flow is well-known in fibre suspensions.

De Gennes[12] has recently demonstrated that this can be understood as a percolation threshold transition. Increased shear will break the structure. The structure does not need to exist in the suspension at rest.

It is worth noting that the stress-strain curve under load cycling of a pile of moist silt has the same main features, except the scale, as a similar curve obtained in a test of a wet paper web. The system behaves in the same way whether made of glass or cellulose fibres.

It may be disturbing to some that we have here a structure which exists without the assumption of any attraction between the particles. Bonds are not necessary for the display of mechanical strength properties.

It is interesting in this connection to note that if a physicist is confronted with a polymeric material with more or less random molecular orientation, he will immediately think in terms of bond structures and try to solve the problem of bond structures in a random system.

It is worth noting that on encountering the problem of a T_g (polymer glass transition temperature) transition the views of these two schools of science become irreconcilable.

The important point is the realisation that the chemist's approach is at best only a pseudo-potential theory.

Research on mechanical properties of paper can broadly be divided into two main sectors: engineering aspects, and studies trying to find the basis of the so-called paper bond.

Typical examples of engineering studies, are research on the bending properties of paper, board, and laminates like corrugated board, and studies of clamped paper panels. The mechanical behaviour of paper rolls has been studied as stressed skin structures. The elasticity theory used in these studies implies that the results apply only to paper at low relative humidity and under loading of short duration. The boundary conditions for the applicability of the models is sometimes incompletely stated.

These studies have been applied for instance, to predict the stiffness of multi-ply boards. A useful list of stiffness parameters for various paper-making materials was published in 1967[13]. Later research essentially dealt with the development of experimental methods to determine stiffness, and some elegant methods have been proposed[14].

In general the theoretical work has had only limited impact on development, and it seems probable that these conditions will prevail. Because of the cheapness and ease of handling, it will probably always be easier to produce a model of paper or board and study it, than to start from theories. Engineering intuition is then the key to development. The tremendous expansion of the application of various types of board is probably due to this very ease of producing prototypes and their testing under realistic conditions. It proceeds at many places simultaneously because it requires few tools. The expansion of the use of corrugated board is to a large extent dependent on innovation of this type.

A very large group of studies has dealt with elucidation of the mechanical properties of paper in terms of the so-called paper bond. Examples of types of such studies are given in Table 1. More than one thousand papers can be fitted into this table. Mechanical strength approaches are then frequently inextricably mixed with speculations about the basis of refining (beating).

In this work a considerable amount of data has been collected. Some of these experimental data, as yet of little practical significance, may eventually become useful. Some techniques have a high degree of elegance.

But much of the published material is of limited general value for various reasons. Frequently the material under study has been incompletely identified. Very often imprecise experimental methods have been used. The lack of statistical treatment of the data, sometimes rather scant, is frequently

flagrant. The boundary conditions for the applicability are often incompletely stated.

Many of the relatively firmly expressed ideas are speculative; few stand up to rigorous scientific analysis; examples of impressive mathematical approaches may reduce to data-fitting; some theories result in interpolation formulae.

Many papers and some of the few handbooks in the field of paper technology repeat the conclusions of such work, sometimes expressed in more definite terms than the original authors. Critical reviews based on extensive scientific background of recent fundamental research results are highly desirable, and some such work has recently appeared.

Chemistry	Wood chemistry
	Polymolecularity
	Chemical additive, binders
Morphology	Fibre structure, fibre defects
	Single fibre strength
	Fibrils, fines
	Exposed surfaces
	Conformability, collapsibility
Relation to Water	Surface potential, surface tension
	Swelling
	Partial solubility
	Hydrogen bonding
Electrochemistry	Polyelectrolytes
	Electrolytes
	Electrokinetics
Optical	Opacity
Engineering	Refining, beating
	Formation
	Consolidation
	Drying conditions

Table 1
Disciplinary analysis of the influences on the mechanical properties of paper

The scientist's motivation for the above type of work is based on a hope, and sometimes a promise, that it is possible to make predictions of a general and perhaps technical significance from a basis of various physico-chemical, and morphological properties of fibres.

In the present author's view, there has often been an overbelief in the carrying capacity of the various approaches. The analysed system is far too complex for many approaches. There are very few, if any, real systems where it has been possible to integrate knowledge of the behaviour of fragments of a system to explain the behavicur of the system as a whole. The introductory part of this paper shows this to be the case for far simpler cases than paper.

The discussed type of `reductionist' research has hardly ever led to any discovery of practical value. The knowledge which has been accumulated has generally been sterile. As was observed by Sir Ernst Chain[15] "all that can be expected from this research direction is that the work will become increasingly expensive and less meaningful". It is certainly possible to collect many publishable data but hard to obtain meaningful results.

As Sir Ernst points out, one must be more aware of mediocrity in this field of research than in many others. The number of publications is no measure of originality.

This does not imply that empirical work to find the technical optimum conditions is not needed. But conscientious work of this type is not necessarily improved by theoretical speculations.

Neither can development of broad models of paper structure be ignored.

One approach which may become of value is based on network structure analysis. Intuitively, it seems that models of paper which are based on configuration concepts might be useful. Stress concentration in such configurations might be important. Recent models of the breakdown of stressed, irregular Tobolsky-type net-works under progressive strain can be mentioned[4]. Such models fit stress-strain experiments where very thin sheets of paper have been shown to break down in discrete steps.

Recent development of mathematical theories for additive and cancellative interacting particle systems may be fruitful[8]. The starting point is a configuration where changes are assumed to occur only at one site at a time (spin systems). Such processes can be described by flip rates: the mathematics is based on Markov chains. The important point is that they focus on fracture processes absent in conventional continuum theories.

Let us assume that the paper industry can afford to support fundamental research related to mechanical properties. The question then is where it should be carried out. If the atomistic point is used as starting point probably the right place is universities with advanced polymer science. If the research should aim at the engineering side the work is probably best carried out in departments of mechanical strength analysis. The important point is that it must be carried out by groups of a critical size to ensure deep scientific analysis and criticism, and that the publications come up to the highest standard. The paper industry is not in a position to afford anything but excellence.

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Discussion following papers given by Prof. B. Steenberg and Mr. I.F. Hendry.

Dr. R.E. Mark, ESPRI, USA

Dr. Steenberg observed in his paper that paper sheet tensile properties (strength and index) fall dramatically for basis weights below about 60 g/m^2. I would like to present here the results of some experiments that bear on this, as I am uncertain that surface effects of the fibres are necessarily entirely reponsible.

The determination of tensile strength depends upon a precise measurement of sheet thickness. We have compared `thickness' measurements made in four different ways (for sheets of 15 and 30 g/m^2).

The four different techniques we used were

a. Digitising scans of a series of photomicrographs of a resin saturated sheet (we had first to find a resin that didn't cause swelling).

b. Measuring with a spring loaded stylus connected to an LVDT.

c. Mercury immersion.

d. Conventional caliper measaurement.

We found that for the lighter sheets all four methods gave different answers, in the order a, b, c, d, whereas for the heavier sheets conventional caliper determination gave an answer substantially higher than the others which were all approximately equal.

Thus it seems possible that the values of tensile strength reported in the literature may be underestimated, relying as they do on caliper determination.

The position is different for the specific tensile strength, the tensile index, since it may be calculated in three separate ways, of which only one depends upon thickness determination.

Firstly, the tensile strength can be divided by the volumetric density: secondly, the breaking force can be divided by the linear density: or thirdly, the breaking force per unit width can be divided by the basis weight.

Of these only the first route depends upon sheet thickness, while the other two do not.

Thus it is necessary to seek an explanation for the reported reduction of tensile index at low basis weights other than the uncertainty in sheet thickness.

A possible physical explanation is as follows.

The distance between fibre bonds is substantially greater in 15 g/m^2 paper than in 60 g/m^2. Thus there is likely to be much more lateral instability in compressed fibres in the lighter sheet than in the heavier. This could certainly be expected to reduce the tensile index, irrespective of any surface effects.

Thus, while fibre surface effects may contribute significantly to the reported reductions in tensile properties of lightweight sheets, it is by no means necessary to assume that these effects are alone responsible.

Prof. B. Steenberg.

Fibre surface effects are manifestations of buckling instabilities of compressed fibres. Results of experiments with fibrous network models reported at the Second Fundamental Research Symposium[4] showed that both compression and tension caused surface fibres to loosen and project out from the sheet. The increase in thicknesss during stressing is well known, for example, in multiwall paper.

Mr. A. de Ruvo, STFI, Sweden

I am interested to see that in your review you have dispensed with the hydrogen bond model of paper structures. This gives me some comfort, though others may be a little frustrated.

Prof. B. Steenberg.

Though I have not included the hydrogen bond model, I do not intend to imply that it is dispensed with. I have only discounted its usefulness in predicting the unknown properties of paper. I doubt neither the hydrogen bond nor its importance. However, I do not see its relevance to practical paper-making, for the reason that the hydrogen bond itself is not amenable to experimentation. The old deuteration experiments on virgin pulp and paper made from it were very useful, but, to my knowledge, no such experiments are being conducted at present. There are lots of calculations being made, but these are not being supported by new experiments, and rely on data from the literature.

Dr. A.H. Nissan, Westvaco, USA.

I have some hitherto unpublished results that may go some way to answering Prof. Steenberg's criticisms.

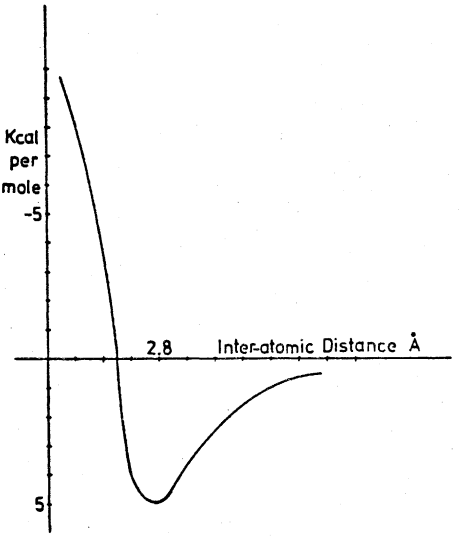

Fig 1

The results come from experiments performed at the Forest Products Research Laboratory, Madison, Wisconsin, by Dr. Von L. Byrd which have been confirmed by Dr. John Glomb, at the Westvaco Research Centre, Covington, Virginia.

Consider the potential energy function for a hydrogen bond, shown in figure 1.

This illustrates that the equilibrium distance, Re, for the O-H-O structure is approximately 2.8-2.9 Å, at which range the bond energy is 4.5 - 5.0 kcal/mole of H-bonds. There is a force constant k_r which measures

how easy it is to rupture the bond, which has been measured frequently over the past twenty years by spectroscopic means. The average value obtained from all these determinations is

$$(1.5\pm0.1)\times10^{4} \text{ dynes/cm.at } 0^{\circ}C.$$

Dr Schuster of the University of Vienna has kindly supplied me with the latest quantum mechanical estimates for the potential based on the two best calculations known to him, from which a value of

$$(1.45\pm0.2)\times10^{4} \text{ dynes/cm}$$

is obtained.

Thus quantum mechanics and spectroscopy may be said to give a combined value for k_r of

$$(1.5\pm0.2)\times10^{4} \text{ dynes/cm}$$

The hydrogen bond theory referred to by Prof. Steenberg may be summarised by the equation

$$E = k_r.N^{1/3}$$

where N is the number of H bonds per cubic centimetre of paper, and E is the Young's modulus.

Now consider the set of curves for cyclic stress-strain loading shown in figure 2.

The total energy absorbed per cycle, TEA, represented by the area under the hysteresis loop, A, can be expressed in terms of energy per unit volume, This is the irreversible energy dissipated in a stress-strain cycle.

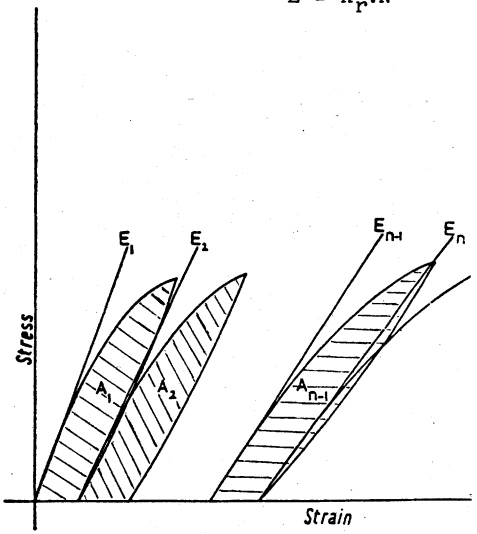

Fig 2

Dividing this number by the energy necessary to break one bond, given above as

4.75 kcal/mole of H-bonds (3.3×10^{-13} ergs/bond),

gives an estimate of the number of hydrogen bonds (M) broken during the cycle. Thus

$$M = TEA/(3.3 \times 10^{-13}) \text{ bonds/cm}^3$$

If the sample is subjected to a number of cycles (n), then the sum of all the TEA values is related to the total number of bonds per cm^3 broken during all the cycles (T) by the expression

$$T = \sum_{i=1}^{n-1} (TEA)_i/(3.3 \times 10^{-13})$$

The initial and final values of Young's modules, E_1 and E_n, are given by the equations

$$E_1 = k_r . N_1^{1/3}$$

$$\text{and } E_n = k_r . (N_1 - T)^{1/3}$$

Since determinations of values of Young's modulus are only accurate to $\pm 3\%$, it is necessary to break as many bonds as possible in order to be able to distinguish between E_1 and E_n.

To achieve this the experimentalists repeated the stress-strain cycles some twenty times at increasing strains. By summing the TEA value from each hysteresis loop they were able to calculate the value of k_r directly from the rheology of paper sheets.

I am indebted to Dr. V.L. Byrd and to Dr. J. Glomb for their permission to quote their experimental results for the values of k_r, namely

(1.25×10^4) dynes/cm and (1.5×10^4) dynes/cm

respectively, with an average value from the seven samples at the two laboratories of $(1.32 \pm 0.2) \times 10^4$ dynes/ cm.

This experiment includes no adjustable parameters and is a truly predictive experiment with exact relationships between the various parameters. We need more work of this kind to confirm the results beyond reasonable doubt and to try to answer the many very intriguing questions in paper rheology.

Prof. B. Steenberg.

My paper is a critical review of the available literature. I am very glad to have seen this information, and look forward to reading about it when published.

Dr. L. Eriksson, STFI, Sweden

I would like to add a comment to Mr. Hendry's paper.

A lot of research is needed before we can hope to achieve any sophistication in quality control and it is my belief that the current proposals for mill-wide control systems from the major suppliers are not appropriate. The full inter-relation between process and product variables must be understood, and while it is not it remains an area demanding research effort.

Mr. I.F. Hendry, Wiggins Teape, UK

I entirely agree. Indeed an earlier speaker asked for suggestions on topics for fruitful research. I suggest that by spending a month in a paper mill any research worker would accumulate so many problems that he wouldn't know where to start.

Mr. A. Ibrahim.

I fully agree that a great deal remains to be done in trying to understand the entire paper making process. However, I do believe that some of the modern control equipment available provides helpful information. We, the control equipment manufacturers, certainly do not believe we have all the answers, but our aim is to provide tools to discover what is happening throughout the process.

Mr. B. Radvan, Wiggins Teape, UK.

I keep thinking of Professor Tabor's remark about respectable and less respectable research. Though his remark referred to research into the physics of the solid and liquid states, I wonder if the same problem doesn't affect the areas covered by Mr. Hendry's paper.

Mr. I.F. Hendry.

I have my own comments to add here. Peter Wrist and I were both up at Cambridge together, and even shared a tutor. Since then Peter has done the `respectable' research, while I have done the `less respectable' work.

Prof. K.I. Ebeling, Helsinki University of Technology

Mr. Hendry, does part of the success of the integrated control system derive from the acute financial conditions which prevailed in the paper making industry throughout the 1960's and 1970's? Under those conditions, even the slightest process improvement, implemented all year round, gave rise to enormous financial returns. Against such a background, it is easy to understand both the rapid penetration of these systems, and the research effort that was mounted to make the sensors reliable.

Dr. H.F. Rance.

I am of the opinion that the development of accurate and reliable sensors was responsible for the rapid spread of control instrumentation.

Mr. I.F. Hendry

Obviously the consideration of a rapid return on investment did play a large part in the acceptance of control technology, but, as Dr. Rance has implied, there was no return on the early substance and moisture gauges, because of the difficulties in making them work. It was only the advent of reliable measurements that allowed the closing of the control loops.

Mr. I.K. Kartovaara, Enso-Gutzeit Oy, Finland.

Practically all quality control is based on the simple calculation of an arithmetic average. This is acceptable provided that the underlying distribution of the variable in question does not change with time. But in general the use of other types of statistics in quality and process measurement should be considered.

Mr. I.F. Hendry

I fully agree. But surely that is already done by modern on-machine gauging systems, where the statistics used are considerably more complex than simple means and standard deviations? Continuously updated analyses of variance, regression, and Fourier analysis are all possible in modern control packages. The widespread use of computers in these applications has made available far more advanced statistics than ever were in the days of hand calculation.

I think these new techniques are used to advantage.

Mr. P. Wrist, The Mead Corporation, USA

I would like to make three general observations on the proceedings so far.

Firstly, on the question of whether or not fundamental research should be directed, we have heard arguments both for and against. My feeling is that while either approach can lead to useful results, some direction is necessary if efficient use is to be made of limited resources. Referring to my own experience, Dr. Wahren described how my fundamental work on table roll drainage was strongly directed with a short term deadline. Similarly, in the second company I worked for, the management's clearly stated intention of being the world's fastest newsprint producer, allied to its policy of posting results daily, ensured that fundamental research was performed only when immediately applicable. And again later, when my research into the factors affecting fibre dispersion uniformity, and sheet formation and structure, was quite general, there was always a specific, known application for the results. This, I believe, focussed my

investigations admirably.

Thus it has been my experience that the chances of new knowledge's finding early application are considerably improved if fundamental research is directed, and if the researcher is kept aware of what the direction is.

Secondly, I want to draw attention to the considerable value of team-work. As Dr. Mardon mentioned, there was a number of research teams working on various aspects of paper manufacturing in the early 1950's, each of which interacted with the others to some extent. I certainly recognise the enormous help I received from others, both within and outside the teams of which I was a member, whether at St. Winifred's, at Baie Comeau, or at Chillicothe. In the case of the development of drainage foils, it was certainly not enough to understand how the suction was created: it was absolutely essential to work alongside other teams, especially in the area of materials development, if use was to be made of the knowledge. The first foils were stainless, and could be used only with plastic wires. The development of ceramic foils for use with wires of any material came only later.

Finally, I think we should all recognise the role of competition in fundamental research. There was a number of Cambridge graduates beginning in industry at the same time as I. One in particular was very active in my areas, and provided me with a lot of competition. He began in Wiggins Teape, followed me to Canada, and later followed me to the USA. In today's proceedings he has already beaten me to the punch. As you may by now have realised, my long standing competitior is Jasper Mardon, whose continued rivalry has provided my work with a tremendous stimulus.

The following contribution was received after the Symposium.

Dr. A.H. Nissan

After the session several colleagues expressed their desire to repeat these experiments and to study other factors. These comments may be of some use to those people wishing to do this.

We found it necessary to take some of the following precautions:

1 Mechanical conditioning: It is necessary to stress the sample to a strain of some 0.1 - 0.2 % and destrain it to zero a few times before the series of loops is measured.

The first rising limb of the first loop is affected by many special factors and is usually not reproducible. (Indeed, as Dr. Corte has pointed out, this limb can take the shape of a J curve, showing strain hardening.)

In practice the first curve is of great importance, but for our purposes it introduces too many transient and irrelevant factors, and should be eliminated. Otherwise E_2 may be found equal to or even exceeding E_1.

2 Conditions should be isothermal: This means a very slow rate of straining and destraining, otherwise temperature (and even humidity) effects will intrude, again making E_2 values unreliable, even exceeding E_1.

In those of our experiments where the loops were taken to high strains, the rate of destraining was maintained so low that it took twenty minutes to complete a loop.

3 Signal to noise ratio: A single loop will rarely produce an effect much larger than a standard deviation in the measurements of E, so we found it necessary to conduct 15 to 20 loops per sample to improve the signal to noise ratio.

4 Protocol: We found the following protocol useful.

Supposing we performed 16 loops, so that we had a collection of measurements of E from the beginning of each loop, ie E_1, E_2, ..., E_{16}, then we formed the following pairs:

$E_1 - E_{16}$, $E_1 - E_{15}$, $E_1 - E_{14}$, $E_1 - E_{13}$, $E_1 - E_{12}$;
$E_2 - E_{16}$, $E_2 - E_{15}$,....,$E_2 - E_{12}$; up to ..,$E_5 - E_{12}$.
Thus we obtained twenty five calculations in five sub-groups.

By calculating the means and standard deviations for all the sub-groups and careful examination of each pair, aberrant results could be detected. These were not discarded. The mean of the twenty five calculations was only slightly affected by gross errors.

By reporting the means, the number of calculations, and the standard deviations, all statistical parameters for later evaluation of consistencies and precision become available.

Before calculations of T can be used for studying fatigue, creep, stress relaxation, etc., the following questions about k_r must be answered (hence the delay in our publication of the results so far obtained).

Would values of k_r obtained rheologically from news-print, paper from semi-chemical pulp, unbleached kraft, bleached board, glassine, and regenerated cellulose show regular deviation from the values obtained spectroscopically and quantum mechanically?

What are the effects of radical changes in the process of making the sheet, for example, press-drying?

What are the effects of physical parameters, temperature and humidity, and of chemical parameters, pH and various ionic concentrations (structure making and structure breaking salts)?

Accurate, precise, and reproducible answers to these questions from many different papers tested in different laboratories will prove far more useful in bridging the gap between the H-bond continuum approach and the discontinuous structuralist methods than the useless polemics on irrelevant issues so far raised.

After all, the two descriptions are complementary, not mutually exclusive.

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