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# THE INTERDEPENDENCE OF PAPER PROPERTIES

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#### Synopsis

The difficulties involved in interrelating paper properties are examined. Qualitative relationships through trends and experience are noted and the special case of quantitative interrelationship among burst, tensile and stretch is described.

More basic considerations such as fibre length, fibre strength, sheet density, fibre orientation and interfibre bonding are individually related to common strength properties and special consideration is given to tearing strength. These more basic considerations are themselves interrelated and an attempt is made at a unification of paper properties through such basic interrelationships. It is suggested that fibre strength, in a broad sense, is of particular importance in such unification.

#### Le rapport des propriétés du papier entre elles

On examine la difficulté d'établir des rapports entre les différentes propriétés du papier. On signale certaines relations qualitatives établies par l'expérience et par l'étude de certaines tendances générales. On traite également le cas particulier de l'interdépendance quantitative des méthodes couramment employées pour évaluer la résistance mécanique du papier, telles que l'éclatement, la résistance à la traction et l'extension. Les propriétés fondamentales, telles que longueur et résistance des fibres, densité de la feuille, l'orientation des fibres et les liaisons entre celles-ci, ont chacune son influence sur les résultats des essais courants cités plus haut. L'auteur parle de la résistance au déchirement en particulier. Ces propriétés fondamentales dépendent également les unes des autres et l'auteur essaie d'unifier les propriétés du papier sur la base de ces rapports fondamentaux. La résistance mécanique des fibres individuelles est considerée comme ayant une grande importance.

#### Die Abhängigkeit der Papiereigenschaften untereinander

Nach einer Untersuchung der bei der Gegenüberstellung der Papiereigenschaften auftretenden Schwierigkeiten wurden die qualitativen Beziehungen aufgezeigt, die durch Tendenz und Erfahrung gegeben sind und besonders die quantitativen Beziehungen zwischen Berstdruck, Reisslänge und Dehnung beschrieben. Die Begriffe der Länge und Festigkeit von Fasern, der Blattdichte, der Faserorientierung und der Zwischenfaserbindung wurden im einzelnen im Bezug auf die Festigkeitseigenschaften beschrieben und besonders eingehend die Durchreissfestigkeit behandelt. Da diese grundlegenden Faktoren untereinander in Beziehung stehen, wurde ein Versuch zur Zusammenfassung der Papiereigenschaften auf dieser Basis unternommen, wofür die Faserfestigkeit von grosser Bedeutung ist.

#### Introduction

THE interdependence of at least the more commonly significant properties of paper is well-known in a qualitative sense to all knowledgeable people engaged in the manufacture and utilisation of paper. The papermaker knows that the required adjustment of one property by some change in the art of processing during manufacture will bring about changes, frequently undesirable, in other properties. The user of the paper is reconciled to the fact that one property may be partially sacrificed in order to attain a high level of another. The sum total of the properties of a sheet of paper for a specific purpose may be regarded therefore as a compromise that is the best attainable within the present limits of the art of manufacture and our general knowledge.

This compromise in a sense is the integration of the various properties of paper. Even within the framework of a given pulp furnish, the permutations and combinations of the various stages of manufacture from head box to calender stack all provide a reasonable degree of flexibility to the papermaker, enabling him to vary certain individual properties over a fair range. This flexibility does not, however, provide the basis for any important improvement in the compromise toward the optimum. Certain other factors in the total manufacturing process from wood to converting operations can, however, affect this compromise very markedly. Modern experience, with the judicious use of mixtures of species of widely differing properties as fibres, has led to the manufacture of grades of paper with distinctly improved compromise of properties. Rapid strides are being made in this area; but, unfortunately, progress is being made largely on an empirical basis. Lack of basic knowledge on the theory of beating and what we are attempting to accomplish during stock preparation and of the mechanisms involved in the action of beaters and refiners is well recognised and requires no further emphasis here. This is true also of beating action on fibres of various species with their differences in dimensions, wall thickness, inherent points of weakness and the like. It would appear to be quite safe to assume that increases in such knowledge must lead to a better compromise in properties, for any given pulp furnish, than that available today.

The increasing use of extraneous chemicals is, of course, largely based on the attainment of this improved compromise. From the older use of clay filler in paper and the like, we have advanced to extraneous interfibre bonding agents and functional surface barrier films for a wide variety of purposes. These are serving largely to improve one or two specific properties with little degradation of other properties and thus serve to raise the level of compromise toward the optimum. Very many examples can be cited in various respects: thus, a very thin layer of an oil insoluble, film-forming material applied to the surface of paper serves to hold out oil, without the necessity of increasing drastically the density of the paper in order to serve the same purpose.

Thus, it is seen that the general rules rightly accepted for the interdependence of the properties of paper can in fact be radically altered with the inclusion of extraneous materials added to the paper and that it is altogether likely that improved stock preparation will create improvements, similar in nature—if not in degree—in the integrated properties for a specific purpose.

The following discussion will be restricted in the main to paper without extraneous additions, however, in order to examine the basic problem of the fibre web itself.

#### Difficulties in assessing relationships among paper properties

As noted above, qualitatively, the interdependence of a number of properties of paper is generally recognised. Trends in the increase or decrease in certain properties resulting from changes in a controllable variable have been well known for many years. The familiar graphs (Fig. 1) of changes in tensile, burst, tear and fold with increasing extents of beating consequently lead to certain qualitative conclusions about the interdependence of these physical strength properties. The most familiar of these is the negative correlation between tensile or burst on the one hand and tear on the other.

33—F.S.P.: i

The positive correlation between tensile or bursting strengths with apparent density, plus the negative correlation of opacity with apparent density, has led to the well-known interdependence between these physical strengths and opacity. Many other instances of such qualitative recognition of interdependence among the properties could be quoted, but they are too well-known to list or to deal with other than by example.





Fig. 1-Change of properties with beating

The question of the interdependence of the properties of paper in a quantitative sense is, however, quite different. Here, the whole literature on paper contains only two or three instances of such information and these will be referred to in some detail below. It is particularly important to examine closely the reasons for the apparent lack of research in an area of such obvious importance as the interdependence of the properties of paper, not only from the theoretical point of view, but obviously also from the standpoint of direct practical application. It would appear that there are two bases in particular for this situation.

One of these is the inherent nature of the physical strength tests commonly adopted by the industry. These tests are designed to yield values representing the resistance of a sheet of paper to stresses of various kinds at the level of the failure of the paper. These tests are necessarily arbitrary in nature and this is recognised in the various conditions laid down for the various so-called standard tests that have been set up in different areas by national technical bodies in various associations. Tensile strength measurements, for example, will differ with the length of span between the clamps. This variation is generally ascribed to the chance of defects and non-uniformities per unit length of the sample strip and this probability will, therefore, increase with increased length of sample. Rate of straining also has an effect, particularly marked at high speeds. In the measurement of bursting strength, the effect of rate of application of load is well known. The level of bursting strength is dependent also on the diameter of the orifice and the properties of the elastic diaphragm used. The endurance folding strength test involves tension, compression and various stresses and strains in shear and the results obtained vary widely with the design of the instrument. The tension employed in the instrument used, for example, strongly influences the results. It is little wonder that different instruments for the measurement of folding endurance may actually rank a series of papers in different orders of merit. The value for stiffness depends greatly on the extent of the bending carried out, since more intensive bending involves the region of plastic flow in contradistinction to elasticity for low load bending. This varies with the instrument used. Reflectance varies considerably with the wave length of the incident light and different instruments may employ different filters. The same holds true for the various opacities deduced from reflectance measurements. Other examples could be quoted further. To this should be added generally that all of these properties must be measured at a standard temperature and relative humidity. although the moisture content of the paper has a different effect in various ranges on different properties. Even the weight of the paper used must be arbitrarily set, since here again variation in basis weight has different effects on different properties.

In addition, it has been increasingly recognised in recent years that a value representing the level of failure of the paper, as a result of the imposition of some type of stress, is not necessarily a representation of the quality of the paper for the intended application. In some cases, quite erroneous conclusions may be drawn from such failure values alone. The best-known example of this lies probably in the use of ultimate tensile load alone, compared with measure-

ments of stress and elongation to obtain work done. It is frequently found that a combination of lower ultimate stress and greater elongation yields a superior paper for specific purposes. The researches carried out during the past fifteen years or so on the prerupture behaviour of paper have served to add tremendously not only to our interpretive knowledge of the structure of paper, but also directly to an increased knowledge of the properties of paper.

The individual properties themselves are very complicated in nature in view of the make-up of paper. Of the physical strength tests, probably tensile strength is the least complicated; even here, we undoubtedly have a complicated mixture of tensile, shear and flexural forces exerted on both fibres and interfibre bonds. When we consider folding endurance, the variety of forces exerted and complexity of the problem are even greater. Much more work is required to make available a clearer picture of what actually occurs as a result of the application of such stresses. Comparable lack of knowledge is encountered in fluid permeability and optical tests.

In view of all of the above, it is apparent that interdependence of paper properties in a quantitative sense cannot be arrived at in the usual sense of direct correlation among these properties, except under such restriction of conditions and arbitrariness of methods as to render any results of little value.

#### Relationships of fundamental characteristics to paper properties

AGAINST this analytical approach, what might be termed an approach involving a synthesis of more fundamental characteristics of paper would seem to be much more desirable and probably much better designed to show up specific areas of lack of knowledge for future research. Paper is a flat, relatively thin web comprised of fibres disposed at various angles essentially in the plane of the sheet and interlaced to a large degree, yielding numerous crossover points at a large proportion of which the fibres may be bonded together. All properties of this web must be reflections of its intimate structure and of the properties of the individual fibres of which it is composed.

Thus, the web must be characterised by the nature of the fibres of which it is composed, their arrangement with respect to one another and the extent to which they are bonded together. This may be classified as follows—

#### Fibre properties

The fibre properties involved may be divided into two classes—(1) fibre dimensions and (2) fibre strength. Of the fibre dimensions, the length has been considered as probably the most important. The axis ratio (that is, the ratio of length to diameter), the cross-sectional shape and fibre wall thickness will

be of importance, particularly in the modulus of flexure of the fibre and are included with the dimensional factors of the fibres. Fibre strength includes not only the tensile strength in the direction of the fibre axis, but also (possibly to a greater degree) the strengths in shear and in flexure and, in general, the integrity of the fibre and absence of gross faults in the fibre denoting zones of incipient failure.

#### Fibre arrangement

Fibre arrangement includes two chief factors—(1) the degree of separation of the fibres in the general thickness dimension of the web and (2) the orientation of the fibres in the plane of the sheet. To reduce complexity, no account is taken here of any angle of orientation to the plane of the sheet or to nonuniformity of distribution of the fibres as evidenced by clumping of fibres and consequent imperfect formation. Another factor in the arrangement of fibres is the amount of coiling or wrinkling of individual fibres, frequently referred to as microcreping.

#### Interfibre bonding

The extent of interfibre bonding basically will be represented by the product of the number of individual bonds and the strength of each individual bond. These factors cannot yet be separated in the present state of our know-ledge, although some advances are being made in this direction. In view of the relationships that have been developed in recent years between bonded areas and scattering of light, we must at present represent the unit of interfibre bonding on the basis of the bonding force per unit area involved at crossover points between fibres, at which locations so-called optical contact is made. This is dealt with again in a later section.

If we list then the main fundamental characteristics of a sheet of paper for consideration of their effects on the properties of paper, we arrive at the following individual factors with no order of relative importance implied—

- 1. Density.
- 2. Fibre dimensions.
- 3. Fibre strength.
- 4. Fibre orientation.
- 5. Interfibre bonding.

It is not suggested in this discussion that this list of more basic characteristics to be viewed in relation to paper properties is necessarily complete, but it would appear that they represent the most important areas for consideration. The paper properties chosen for examination in the light of these more basic factors are as follows—

Strength	Optical	Permeability
Tensile	Opacity	Air permeability
Burst		Water permeability
Tear		Oil permeability
Fold		
Rigidity		

Interrelationships among these paper properties and the five more fundamental characteristics are discussed below for each of the latter. In examining the state of our knowledge in this area, we might classify the available information broadly into four groups—(1) where quantitative relationships have been measured, (2) where relatively strong correlations have been set up both through laboratory investigations and on the basis of many years of papermaking and paper evaluation experience, (3) where a reasonably good case can be made for relationship by inference and (4) where no reason for any correlation can be found on the basis of cause and effect reasoning. It is of particular interest to note that none of these possible relationships can safely be placed in the last-named category. This is simply a result of the fact that all of these paper properties are interrelated to a greater or lesser degree; furthermore, they are based on the common denominator of a combination of the five basic characteristics set out. The effects of the latter are set out individually in the succeeding sections.

#### Density

OF the five fundamental characteristics here considered, the apparent density of the paper would appear to be the most important and, in fact, the dominant factor. It is well, therefore, to evaluate the basic considerations governing the level of density of a sheet of paper and its interrelationship with the other four fundamental characteristics under discussion prior to consideration of its effect on the various paper properties.

The manufacture of a sheet of paper following pulping, bleaching and stock preparation consists essentially of the removal of water from the fibres by various means—drainage, pressing and evaporation. During these water removal procedures, the fibres are brought into closer juxtaposition in the thickness dimension, so that the dry weight of fibres per unit of wet thickness increases continuously. For any given fibre dimensions, therefore, a given number of fibres representing the final basis weight of the paper are continuously being brought closer together. The forces bringing about this thickness contraction include surface tension, interfibre adhesion, the extraneously applied mechanical forces of vacuum and pressure, interfibre bonding, area shrinkage of the fibres and the coiling and twisting of individual fibres during the last stages of the drying.

The contraction of the web in the thickness dimension begins at a very early stage in manufacture, long before drainage is complete on the wire. Up to the point that sufficient water is still present to fill completely the spaces among the fibres in the web, surface tension forces are very low, since the interfacial area is merely that of the two surface areas of the web. When, however, drainage is advanced to the point when air begins to form an appreciable proportion of the medium in which the fibres are suspended, the water/air interfacial area increases rapidly as does also the resultant force of surface tension. This begins to occur at a solids content of only about 11-12 per cent, that is, well back along the forming wire of the papermachine. It has been shown by Lyne and Gallay<sup>(1)</sup> that the decrease of thickness of the web is very marked following this point and that surface tension is the force bringing this about. Similar contraction is found for non-cellulosic fibre webs, including, for example, glass and various synthetic resins. Lyne and Gallay (Fig. 2) found further that these surface tension forces decrease rapidly after 20-22 per cent solids, when a plateau is reached for a short interval in the graph of web caliper against solids in the web. They showed that some form of interfibre adhesion then continues as a contractive force in the regular progression of decreasing caliper during further loss of water.

Vacuum at the end of the forming wire and particularly the passage through the nips of wet presses will obviously mechanically squeeze these fibres together. Eventually, after the removal of free water on the surface of the fibres and from the interstices between fibres, the fibres themselves shrink owing to loss of water within the fibres. The volume shrinkage resulting is mainly in the cross-section, which is essentially normal to the plane of the web and thus less space is occupied by the fibres in the thickness dimension of the web. During this time, surface tension forces exerted between adjacent fibres at crossover points become very great in magnitude as pointed out by Campbell<sup>(2)</sup> in his well-accepted theory, so that the distance between these adjacent surfaces may be sufficiently decreased to enable hydrogen bonding to occur.

The surface tension forces operating may, in general, be assumed constant, except in such special cases in which surface active agents have been deliberately added to the stock for specialised purposes or when poorly washed stock is used, with consequent retention of surface active materials produced during the pulping operation. In such special cases, it is well known that a low density sheet results, mainly in all probability as a result of the drastic reduction in contracting forces, possibly also through the obviation of hydrogen bonding as a result of the masking or reacting of polar groupings on the surface of the fibres with the surfactant. With surface tension constant and under the assumption also that polar hydroxyl groupings are exposed and



Fig. 2—Change of thickness with solids in the wet web (Lyne and Gallay)

available for hydrogen bonding, the differences in densities in various papers made in the same way must be ascribed to the resistance offered by the fibres themselves to these compacting forces.

It is apparent that the governing factor here is the modulus of flexure of the wet fibre. A fibre with a low modulus and that is plastic rather than elastic in nature will offer less resistance to the compacting forces caused by surface tension and will retain deformations brought about by mechanical pressing. This will lead to a higher density. Furthermore, this degree of plasticity will largely govern the extent of interweaving during deposition, hence the number of potential bonding areas. The pulp fibre is relatively highly crystalline and as such is stiff and springy. Such fibres, on deposition from dilute suspension on the wire mesh, will tend toward a brush-heap type of mat with a minimum number of potential contacts for bonding. The high degree of elasticity will result in a high degree of springback after compression in the nip of the wet press.

Density is therefore governed essentially by the degree of plasticity developed in the wet fibres when introduced on to the papermachine. This depends in turn, for any given type of fibre, on the amount and type of beating carried out and there appears no doubt that the most important aim of beating must be the introduction of this plasticity into the fibre in preparation for the formation of the web. Unfortunately, however, beating brings about effects other than fibre swelling and the enhancement of fibre plasticity. Beating, after an initial stage, results in the fibre shortening, which is well known, also undoubtedly in various types of fibre damage. Thus, ordinary stock preparation changes not only density, but fibre length and fibre strength as well. The proportions of each undoubtedly change with the type of beater or refiner used, the conditions under which the stock preparation is carried out and the extent of the beating in terms of energy applied. Harrison<sup>(3)</sup> showed many years ago (and it has been well recognised since) that a relatively high degree of strength is attained at an early stage in the beating process before any evidence is seen of fibrillation or other apparent damage to the fibre. The same holds true of opacity. Nordman<sup>(4)</sup> found a higher rate of decrease in specific scattering coefficient at early stages of beating. The rate of response of the fibre to beating in terms of added plasticity and flexibility, the ability of the fibres to undergo the mechanical stresses of beating with a minimum of cutting and other damage each depend on the damage introduced in chipping, the method and conditions of cooking and the degree of damage during bleaching, so that a definite type and amount of beating will produce results dependent on the previous history of the fibre. In this connection, the work of Stone and Nickerson<sup>(5)</sup> showing quantitatively the drastic degradation in the properties of sheets prepared from pulp that had been made by the sulphite process from wood previously subjected to compression parallel to the grain is of particular interest. Forgacs<sup>(6)</sup> has emphasised the importance of evenly spaced irregularities along the fibre (which he terms nodes) and has shown that these constitute points of weakness in the fibre, which are further developed, if the sulphite process has been used for fibre isolation, by the action of beating.

This is apparently related directly to the very flexible fibres examined by Forgacs, Robertson and Mason,<sup>(7)</sup> who showed the segmented movement of fibres in a shear field. Damage of this type and that due to overbleaching and other causes have been shown regularly, in the course of investigation in the author's laboratory, to result in a higher sheet density. Here, however, the usual higher tensile and bursting strengths mormally associated with higher density are not obtained and the fibre apparently conforms more readily to the compacting forces exerted during sheetmaking partly through these points of weakness, which are developed further during beating and which enter strongly into the lower strength levels obtained in the sheet.

The flexibility of the wet fibre is strongly influenced by fibre dimensions. Thick-walled fibres possess greater rigidity, hence yield webs of lower density. Flexibility increases with fibre length,<sup>(7)</sup> with consequent ease of compaction and higher density. High axis ratio (length to diameter) in the fibre tends toward greater sheet density.

In examining the effect of density on the properties of paper, it is particularly noteworthy that increase of density by dry calendering has little effect on strength properties. It would thus appear that the structure of the sheet as set by the fibre arrangement and degree of bonding attained during the removal of moisture from the web is not materially influenced by subsequent further compaction. In the same sense, there is little change in opacity with machine calendering. Permeability properties, as might be expected, are affected to a greater degree. It must be emphasised, therefore, that in a consideration of the relationship of the density of paper to various other fundamental characteristics and to the various physical properties of paper, only the uncalendered density should be used.

Although there are apparently no direct quantitative correlations available, there is no doubt that there is a strong relationship between density and extent of interfibre bonding. When bonding is essentially obviated by masking available groupings for bonding, the density is decreased markedly even if an ample degree of stock preparation has been introduced. Thus, the addition of a mineral oil to the fibre surface brings about a large decrease in density. Basically, an increase in density is largely due to bonding, since increased wet flexibility of the fibres results in the creation of a larger number of more closely positioned areas between fibres and subsequent bonding during the latter stages of drying maintains the fibres within a smaller volume. The density is therefore a co-resultant with bonding of the degree of wet plasticity of the fibres, assuming the availability of hydroxyl groups on the fibre surface for bonding.

Particular emphasis has been placed on density in its relationship to wet

fibre properties and to bonding, since, among the various basic characteristics of the web, it would appear to be particularly important in that it is a dominant representation of the structure. The relationship of density to commonly measured paper properties is discussed below, though due caution must be exercised in examining conclusions drawn from various researches reported. particularly in view of one complicating factor. Variations in density are brought about generally, both on a commercial scale and in the laboratory, by some form of beating and refining. Unfortunately, as noted earlier. different methods and conditions of stock preparation bring in their train different degrees of fibre breakage and damage to fibres without overt breakage. It is therefore manifestly impossible to isolate the effect of density alone in such cases and conclusions must be drawn with some care in relating density with paper properties, particularly if results from various laboratories and mills are compared. There is, furthermore, a considerable difference between handsheets and machine-made paper, particularly when the handsheet is dried against a solid metal plate.

In general, it is recognised that the tensile strength increases with increased density of the sheet. Doughty<sup>(8)</sup> noted that strength development is in the main due to the production of denser sheets and stated that this approached a power function—that is, the strength increases as some power of the density. He concluded that the first portion of beating had its effect mainly in an altered surface condition of the fibre, but that subsequent strength increase was due solely to increase in density and that this portion of strength increase could be duplicated by wet pressing instead of beating. Clark<sup>(9)</sup> has stated that tensile strength is linearly proportional to density.

Bursting strength also is well known to increase with increasing sheet density. In this case, too, Clark<sup>(9)</sup> has concluded that bursting strength increases in linear proportion to density. Since bursting strength is a complex function of the tensile strength and the stretch of the sheet, a relationship that will be examined closely in a later section of this discussion, it would appear to be of particular interest that the density is reported to be linearly proportional to both the tensile and bursting strengths.

It is everyday experience also that in general the tearing strength varies inversely as the density, after an initial period during which (with some pulps at least) an increase in tearing strength is noted with the first portion of strength development through stock preparation.  $Clark^{(9)}$  has stated that, following such an initial period when a maximum of tearing strength is obtained, the tearing strength is linearly proportional to the inverse of the density. Tear is a localised stress and the factors entering into tear are examined more closely in a succeeding section.

Folding strength is a very complicated function involving, among other factors, tension, compression, flexure and an important dependence on the time factor. During the course of the typical beater curve, folding strength increases markedly and then decreases, whilst the density continues to increase steadily. Generally speaking, the factors on which fold depend are so varied and complex that no definite conclusions relating folding strength and density can be drawn from available data in the literature.

The rigidity of a sheet of paper is well known to vary with the density. Smith<sup>(10)</sup> has noted that Young's modulus is proportional to density and therefore that the rigidity should be linearly proportional to the density for a constant thickness. Clark has concluded, however, that rigidity is proportional to the square root of the density. The effect of density must be simply that of involving a greater number of fibres per cross-section to resist the action of a deflecting force when the thickness is held constant. When the basis weight is constant, however, the rigidity varies as the square of the thickness. Thus, the same number of fibres disposed to a greater bulk or lesser density produce a structure of much greater rigidity. This is connected with the more general relationship frequently found for a variety of materials including paper, namely, that the rigidity increases as the cube of the thickness when density is held constant.

Density is well known to be closely related to opacity and an increase in density is associated with decreased opacity. The end point of this is the approach to glassine, in which the enhanced transparency is a combination of greatly increased density together with fibre shortening and debris.

Density is obviously associated also with air permeability. Doughty<sup>(11)</sup> noted that porosity decreased markedly with increase in density, then Seborg, Doughty and Baird<sup>(12)</sup> noted that the equivalent pore radius, a typical structural property, was consistently related to density over a wide range of papers of the same weight and different thickness. These workers found also that the logarithm of the rate of air transmission through paper was linearly proportional to the inverse of the density. Lane<sup>(13)</sup> concluded that the logarithm of the air or oil permeability is proportional to the freeness of the pulp, if freeness is the sole variant. Bell<sup>(14)</sup> and Corte<sup>(15)</sup> both note the dependence of air porosity on closeness of packing of the fibres and agree on a close relationship between air and oil permeability. Albert<sup>(16)</sup> found that air porosity over a wide range is linearly related to oil permeability divided by the square of the sheet thickness. The case of water permeability is complicated by swelling of the fibres and no direct information appears to be available on this matter. Arlov<sup>(17)</sup> found that in unsized paper, as might be expected, the water absorption rate decreased with increasing sheet density. With sized paper, on the other hand, he found the converse to be the case; the water absorption rate increased with increasing sheet density. The mechanism of water transfer would appear to be markedly different in the two cases.

#### Fibre dimensions

FIBRE length will obviously play an important part in the structure of the web. The number of 'free ends' of fibre per unit weight of fibre will obviously be much greater with short fibres, therefore the continuity of the mesh may be expected to decrease, even if the spacing of interfibre bonds remained the same with short fibres as with longer fibres. It is to be expected also that the amount of interweaving and consequently the frictional resistance to pulling apart of fibres will be reduced with shorter fibres. A distinction must be made with respect to a mixture of 'shorter and longer fibres' brought about by the cutting action within one species of fibre and a mixture of naturally shorter and longer fibres originating from two distinct species, for example, hardwoods and softwoods. In the latter case, morphological characteristics beyond length are involved. These include wall thickness and axis ratio and may require for consideration such matters as different moduli of flexure and widely differing tendencies to collapse in cross-section. The potential difference in behaviour between a fibre essentially circular in section and one resembling a flat ribbon is apparent in the matter of wet flexibility leading to level of sheet density, also in the area of potential bonding.

In a comparison of various pulps,  $Clark^{(9)}$  found that fibre length was not an appreciable factor in the sheet density obtained. He found<sup>(9)</sup> that the tensile strength of the sheet varied as the square root of the weighted average fibre length by weight. Doughty,<sup>(18)</sup> working with fractionated pulp and under different conditions of sheetmaking so as to yield different sheet densities, found that—(1) at constant density, fibre length had little effect on tensile and (2) using a definite wet pressure, the shorter fibres gave a higher density and stronger sheet. It would appear from this work that the effect of fibre length on tensile strength is through sheet density and that the density—therefore the strength of the sheet—increases with increasing fibre length. This is readily visualised through the related wet flexibility.

In general, tensile and bursting strengths show similar trends and it would therefore be expected that bursting strength also increases with fibre length. Clark<sup>(9)</sup> concluded that the burst increases linearly with the weighted average fibre length by weight. It might be noted here that it appears difficult to reconcile this relationship with that previously noted by the same author, that the tensile varies linearly with the square root of the weighted average fibre length by weight. Tensile and burst are related through stretch, as will be discussed in a succeeding section, and this relationship does not appear compatible with the two quantitative relationships under discussion. It might be safer therefore merely to note that tensile and bursting strengths increase with fibre length.

It is well known that tearing strength is closely dependent on fibre length. Clark<sup>(9)</sup> has concluded that the tearing strength is linearly proportional to the square root of the cube of the weighted average fibre length. From a theoretical study,<sup>(19)</sup> it has elsewhere been concluded that the tearing strength should be linearly related to the fibre length.

The fibre length is a very important factor also in folding strength; Goldsmith and Higgins<sup>(20)</sup> have described this dependence as critical. Since the various factors entering into folding strength—for example, tensile, stretch, compression—are all related to fibre length, the strong positive relationship between folding strength and fibre length is to be expected. The sharp decrease in folding strength invariably noted during the later stages of beating can probably be attributed largely to fibre shortening and associated sensitivity of the folding strength, as well as to loss in stretch.

The rigidity of a sheet is related also to fibre length, but various other factors must be taken into account. Clark<sup>(9)</sup> found that the rigidity factor was linearly proportional to the square root of the weighted average fibre length, when the degree of bonding and fibre strength are kept constant. As noted earlier, the rigidity of a sheet is very sensitive to the bulk factor—for example, rigidity varies as the square of the thickness for constant weight. The density of a sheet is closely related to the degree of bonding and fibre strength and it is important to note that this must be kept constant in order to obtain a positive correlation between rigidity and fibre length; otherwise, increased density must result in a decrease in rigidity.

Fibre length is related to opacity through two factors.  $Maass^{(21)}$  has shown that the opacity of a sheet increases with decrease in fibre length. Parsons<sup>(22)</sup> reached the same conclusion, stating that fractions of shorter fibre length have higher scattering power and that the latter is a linear function of the total surface area involved. Parsons showed also that mixtures of fibre fractions are additive in this regard. It should be noted, however, that the density of a sheet decreases with decreased fibre length and, since opacity increases with reduced density, at least part of the effect of fibre length on opacity may be through change in density of the sheet.

Fibre length bears a relationship also to the permeability of a sheet to various fluids. Doughty,<sup>(18)</sup> for example, has noted that, for any given density, the air permeability of a sheet increases markedly with increased fibre length. Carson<sup>(23)</sup> found that air permeability is inversely proportional to the thick-

ness of the sheet (at constant density). He concluded that the length of the tortuous path through the sheet was of the order of one hundred times the thickness. The cross-sectional shape of these passages are important and thus the cross-sectional shape of the fibre has an important bearing on fluid permeability.

A further discussion of the effect of other dimensional factors in addition to length is given in the general discussion section of this paper. The wet flexibility of fibres varies with the axis ratio and therefore short, thick-walled fibres will show a lesser degree of flexibility. As a result, such fibres will produce sheets of lower density, hence lower tensile and burst, higher tear and opacity. In addition to this, the response to beating in terms of fibre damage and breakage varies with these dimensions, as well as with certain morphological factors in such fibres. This, in turn, has a corresponding effect on the various physical properties.

#### Fibre strength

In view of the very laborious procedure that would be required to measure directly the tensile strength of an individual fibre and the very large number of such measurements that would probably be required to characterise the fibres of one pulp, because of obvious broad variations from fibre to fibre, very little information is available directly on this fibre property. The zero-span test as devised by Hoffman Jacobsen<sup>(24)</sup> and further developed by Clark<sup>(25)</sup> has proved useful in some instances for the measurement of strength losses by degradation, but cannot be regarded as more than an arbitrary indication of tensile strength. It is well known that the type of pulping procedure used to defiberise wood results in fibres of widely different strengths, but the nature of this difference is still not understood. It has been suggested, for example, by Jayme and von Koeppen,<sup>(26)</sup> that the lower strength shown by a sulphite pulp in comparison with a kraft pulp lies in the weaker interfibre bonding in the sulphite sheet, resulting in turn from a lower degree of polymerisation of the carbohydrate at the surface of the fibre. On the other hand, the recent work by Forgacs<sup>(6)</sup> on the nodes of sulphite pulp fibres and their relationship to probable points of weakness along the fibres is strongly indicative of the strength of the fibre itself being related to the strength of the sheet. Such points of weakness are clearly related also to the wet flexibility of the fibres, hence to the density of the corresponding sheets.

It is well known that various degrading factors affect all the properties of the sheet—for example, Stone and Nickerson<sup>(5)</sup> have shown that the degradation brought about by compressing wood parallel to the fibres prior to pulping results in a general lowering of all strength properties. It is common knowledge that overcooking, overbleaching, exposure to mineral acids and other such forms of degradation result in decrease in sheet strength. It should be emphasised that this does not constitute a shift of values as obtained in beating, during which, for example, the tearing strength is decreased while the tensile and bursting strengths increase, but rather a general degradation of all the strength properties concerned. These forms of degradation produce also a higher density in the sheet, thus the general relationship between tensile or bursting strength with density is seriously altered. It is not possible with present knowledge to state definitely whether this is related to decreased fibre strength, decreased interfibre bonding strength or both; in most of these cases, the fibre strength is likely to be more seriously involved.

When paper fails as a result of some applied stress—for example, tensile, burst, tear-failure must occur as a result of bond breakage or a combination of the breakage of fibres together with pulling apart at interfibre bonds. It was formerly rather tacitly assumed that interfibre bonding was by far the more important of these two factors. More recently, however, this view has changed considerably: Graham<sup>(27)</sup> suggested that, for many stronger papers, the rupture value begins to depend on fibre strength after bonding has reached a saturation value. He suggested that fibre breakage was much more important in tension failure than had previously been recognised and that frequently the sheet strength was limited by the fibre strength rather than by the strength of the interfibre bonds. Van den Akker, Lathrop, Voelker and Dearth<sup>(28)</sup> have made an outstanding contribution to this problem by actual measurement of the relative proportions of fibres broken and pulled out after failure due to applied stress. They found that more fibres were ruptured than pulled out in a tensile break and that this differential increased with the amount of beating. Following failure by tear, they found very much more rupture than pull-out and this differential also increased with the amount of beating used. Increased tensile strength afforded through the use of an extraneous bonding agent was found to increase breakage and the use of a surfactant that lowered the sheet strength resulted in relatively little breakage of the fibres in a tensile test. Rather more fibre breakage was noted with longer fibres. It is apparent from the results obtained by Van den Akker et al. that both bonding failure and fibre breakage are involved and that the significance and magnitude of the factor of fibre strength is much greater than had hitherto been realised. The possibility has been noted by Gallay<sup>(29)</sup> that failure in some cases may take place within the fibre adjacent to a bonded area as a result of damage previously suffered by the fibre during processing.

Folding strength has been noted, in many instances, as particularly sensitive to a diminution of fibre strength; much more so than are tensile or

burst. This may have some relationship to decrease in toughness or embrittlement of the fibres, as well as to incipient zones of weakness along the fibres, which may make them more sensitive to the severe flexural stresses involved in the folding endurance test.

Further reference is made to the particular importance of fibre strength on tear in a later section dealing separately with tearing strength.

#### Orientation

A SHEET of paper may vary in orientation of fibres from an extremely high degree of orientation in the machine-direction to a virtually 'square sheet' with a high degree of randomness of orientation. Generally speaking, in ordinary papers, a pronounced preference in the machine-direction is involved.

The effect of this on tensile strength is, of course, well known and the ratio of the higher machine-direction tensile strength to the cross-direction tensile strength is one of the important characteristics of paper. The tensions applied on the papermachine during manufacture have a pronounced influence on these values, that is, the draws on the machine-direction tensile strength and the tightness of felts on the cross-direction tensile strength. It has been shown by Steenberg<sup>(30)</sup> that this is not due solely to their influence on fibre orientation and it is suspected that this phenomenon may involve an orientation of fibrils or bond orientation. Otherwise, the comparison of tensile strengths at various angles might be taken as a rough measure of the corresponding proportion of fibres oriented at those angles.

The bursting strength, although not a directional test, is affected strongly by orientation through stretch. Stretch is reduced in any direction by the stress applied during drying and the stretch is therefore least in the machinedirection. The bursting test demands an expansion of the paper and the low stretch of the machine-direction is important in the level at which the final rupture occurs. Before the burst can actually occur, the strongest line must be broken and thus the line of burst is usually in the cross-direction. It may be generalised that the bursting strength essentially parallels the tensile strength in which the paper has the least stretch, which is normally the strongest direction of the paper.

Tearing strength is strongly dependent on fibre orientation and machinedirection and cross-direction tearing strengths are different to such an extent that tear trials by hand are commonly used to determine the machinedirection of a sheet, should other evidence not be obvious. Since a large part of the work in tearing involves fibre breakage, it is apparent that the resistance will be very much less if the tear line is parallel to the direction of greater

34—ғ.ѕ.р.: і

orientation, that is, when fewer fibres must be broken in order for the failure line to proceed.

Generally speaking, machine-direction folding endurance (with the axis of fold in the cross-direction) is greater than the folding endurance in the cross-direction (with the axis of fold in the machine-direction). This would generally be ascribed to a greater tensile and compressive strength in the corresponding direction, but the folding endurance test is a complicated one involving several factors and it is understandable that the reverse to the above generality is sometimes encountered.

Rigidity varies with the degree of orientation and this relationship is commonly used by the paper converter or user in determining the direction of cutting of smaller sized sheets for a particular application. Rigidity is much greater in the direction of orientation, involving the modulus of flexure of a greater number of fibres.

#### Interfibre bonding

AFTER fibre deposition and during the process of papermaking, each fibre is in relatively close contact with many others and each crossover region represents a potential area of interfibre bonding. It is generally accepted and well documented that these are hydrogen bonds. The actual total amount of bonding will be the product of the number of bonds per unit area and the total area of bonding. These in turn will depend on the number of available hydroxyl groups on the fibre surfaces at each crossover region and on the interfibre distance. Reduction in the number of available polar groupings-for example, by esterification or by masking with an oil film-is known to reduce bonding drastically. The required intermolecular distance of the order of several Ångström units is brought about by the action of powerful surface tension forces during drying. Reduction of these surface tension forces by surfactants is known to reduce bonding. Hydrogen bonding proceeds through a water medium and the removal of water after deposition, but prior to drying, by freeze-drying or by exchange for non-polar solvents, obviates bonding and results in a weak, bulky sheet. As noted in an earlier section, resistance to pulling together of the fibres to the close contact required is provided by the elasticity and rigidity of the fibres and the process of beating introduces sufficient plasticity into the wet fibres to facilitate this pulling together and subsequent interfibre bonding. Corte and Schaschek<sup>(31)</sup> have suggested that the tension applied during drying on the papermachine assists in bringing closer together those fibres mainly involved in this stretch.

The question of the strength of the bond is of considerable interest. Nordman, Gustafsson and  $Olofsson^{(32)}$  found, for any given pulp, that the

amount of energy required to increase the specific scattering coefficient by 1 unit (that is, to separate a definite amount of previously bonded area) was a constant independent of all the usual sheetmaking variables such as beating, wet pressing, orientation, tension during drying and basis weight. This appears therefore to be a pulp characteristic and is termed *bond strength value* (BSV) by these workers. Since it is unaffected by the various factors entering into the amount of bonded area produced, it represents apparently either a measure of the strength of the elemental bond or the number of such elemental bonds in a unit area of interfibre bonding. It is interesting to note that corroborative correlations were found—for example, kraft pulp had a higher BSV than had sulphite and increased pentosan content led to a higher value of bond strength.

The relationships between interfibre bonding and tensile and bursting strengths are apparent, since interfibre bonding is responsible, in the first instance, for strength. It is logical to assume that these strength values increase with bonding until the saturation value for the latter has been reached. Beyond this point, the limiting factor is the strength of the fibres themselves. Non-uniform stress distribution undoubtedly plays an important part in the process of failure. The concept of Rance<sup>(33, 35)</sup> advanced some years ago. which suggested progressive breakage of interfibre bonds with increased straining, has received excellent corroboration more recently in the progressive accompanying changes in opacity described below. Depending on stress distribution, this pulling apart of interfibre bonds increasingly brings into play the factor of fibre strength. In the case of bursting strength, for which stretch is of considerable importance, increased bonding over a broad range still increases the bursting strength, despite the accompanying decrease in stretch. It is noted that, generally speaking, the burst value begins to decrease with excessive beating while tensile strength is still increasing.

The tearing strength in general varies inversely as the amount of interfibre bonding. Frequently, during the first portion of a beating curve, the tear increases markedly and this is generally explained on the basis of increased frictional resistance to pulling fibres out of the paper with some interfibre bonding introduced. With more bonding, however, fibres are ruptured, involving less work than pulling out and the tearing strength shows a continuous decrease. Further reference is made to this relationship in a later section.

The folding strength increases markedly with increase in interfibre bonding along the beating curve over a wide range. The subsequent decrease in folding strength may be a reflection of greatly decreased stretch and other changes in elasticity factors in the sheet, as well as of increase in fibre damage. It appears likely that the accompanying fibre shortening, to which the folding test is particularly sensitive, plays a part in bringing about this decrease. Mason<sup>(34)</sup> has suggested that high folding strength is associated with a high relaxation rate and a relatively high primary creep in the sheet, so that, on repeated folding, localised stresses and strains do not exceed their failure value. Rance<sup>(35)</sup> was unable to find, however, any evidence substantiating such a relationship between folding strength and recoverable plastic flow. On the other hand, Rance found (Fig. 3) a definite correlation between folding strength and the elastic flexibility of the sheet. The elastic flexibility was obtained either by measurement from the stress/strain curve with a correction



Fig. 3—Relationship of folding strength to elastic flexibility (Rance)

based on the type of folding endurance instrument used or by calculation from independent determinations of rigidity and tensile strength. Good agreement was obtained between the two methods. It is not possible to sort out these various factors with present knowledge, in the interpretation of the course of folding strength with beating.

Rigidity increases with interfibre bonding to a maximum, then shows a continuous decrease. As in the case of tear, a certain amount of bonding prevents fibre movement and thus enhances rigidity. At the same time, however, density continuously decreases and the rigidity is known to decrease as the square of the density for a given weight. These two opposing factors result in

a maximum in rigidity at an early stage in bonding. In the same general sense, Van den Akker<sup>(36)</sup> has suggested that rigidity in thin papers is largely a matter of the stiffness of individual fibres, whereas rigidity in thick papers is more a matter of extent of fibre bonding.

The relationship between bonding area and opacity has received considerable attention and the results obtained are of wide significance in the general problem of paper structure. The method developed by Parsons<sup>(22, 37)</sup> for the determination of bonded area involved the measurement of total surface area and the scattering coefficients of bonded and unbonded sheets. Total surface area was measured on the slush pulp by the silvering technique and unbonded sheets were produced by solvent exchange prior to drying. It was then assumed that the ratio between total surface area and the unbonded area in the normal sheet (dried from a water medium) was the same as the ratio between the scattering coefficient of the unbonded sheet and the scattering coefficient of the normal sheet. From this, the bonded area is obtained by simple calculation. The specific scattering coefficient is thus a linear function of the specific surface area of the sheet. Ratliff<sup>(38)</sup> confirmed this relationship, then compared bonded area with area in optical contact: he concluded that the latter is a function, if not a direct measure, of bonded area. It should be noted, however, that hydrogen bonding involves a fibre separation of only several Ångström units, whereas optical contact still holds up to a large fraction of the wavelength of light (several thousand Ångström units). Nordman<sup>(39)</sup> has discussed this matter at some length, including a review of the work of Haselton<sup>(40)</sup> using a nitrogen adsorption method; he noted that this and other uncertainties exist in equating bonded area with loss in scattering power. Nevertheless, the relationship between scattering power and bonded area must, in the light of other evidence, be considered broadly acceptable and useful and as offering a relatively simple and very valuable tool for further investigation. Nordman has noted the remarkable agreement between the number of bonds per unit volume being subjected to stress during a stress/strain cycle as calculated on a mathematical basis by Nissan<sup>(41)</sup> and the same values derived from his measurements of changes in light scattering.

Ingmanson and Thode<sup>(42)</sup> have corroborated the general validity of a linear relationship between scattering coefficient and surface area of fibres in the web, but have criticised the use of drying from non-aqueous liquids to obtain the total area of unbonded fibres, particularly in view of the lesser shrinkage of the fibres with such a technique and consequently the higher scattering coefficient. They suggest, instead, the extrapolation of the relationship between scattering coefficient and tensile strength to zero tensile, using unbeaten pulp with mild wet pressing. They emphasise the ease and utility of

using area of interfibre bonding for correlation with strength properties. It was concluded from this work that the total dry fibre area available for bonding remains constant with beating and is unaffected by fines or changes in the fibre structure. Swanson and Steber<sup>(43)</sup> concluded from their bonded area measurements that beating brings about a change in bond density within the area of interfibre bonding.

It has been shown by Nordman, Gustafsson and Olofsson<sup>(44)</sup> that the stress/strain behaviour of paper is closely paralleled by corresponding changes in reflectance. Increase in light scattering begins at about the strain value past the Hookean portion of the curve and continues with further strain. The increase in scattering, on repeating a cycle to the same percentage of strain, is small as is also the energy loss. In this connection, it is interesting to note that Nordman concludes that the increase in free area obtained as shown by opacity was not recoverable, which means that broken bonds did not re-form. Nissan<sup>(41)</sup> assumed recovery of bonding on destraining. It has been shown also that unbonded sheets made by solvent exchange increase greatly in strength on standing in an atmosphere of high relative humidity. This is a point of considerable theoretical interest, since fibres are probably under a condition of strain in the bonded web and the release of such a strain must separate two previously bonded fibres by more than about half of the wavelength of light in order to destroy optical contact, hence produce increased scattering. The conditions under which such a bond would re-form must involve considerations of the plasticity of the fibres as governed by moisture content as well as by further strains applied.

Parsons<sup>(37)</sup> obtained a definite correlation between strength and bonded area. Ratliff<sup>(38)</sup> reported that the burst and tensile strengths over a wide range increase linearly with bonded area. A rapid increase in bonded area was noted in the early stage of beating and eventually it was considered that bonding strength approached fibre strength. The more recent change in view about the importance of fibre strength has already been mentioned.

Forman<sup>(45)</sup> has shown that the density, burst, tensile and tear are all linearly related to the logarithm of the beating time. Since bonded area varies linearly with the beating time itself, it could then be shown that these physical properties varied linearly with the logarithm of the bonded area. These interrelationships hold true over a wide range of stock preparation. The extent of parallelism between bonded area and density is of particular interest in view of the ease of measurement of density as a basic characteristic of the sheet. In this connection, it has recently been reported by Chase<sup>(46)</sup> that the increase in density during drying, presumably therefore the increase also in interfibre bonding, closely parallels the increase in tensile and bursting strengths.

#### Quantitative relationships among paper properties

In the above section, an examination has been made of the effect of certain fundamental characteristics on each of a number of common paper properties. If we consider direct relationships among these paper properties,



*Fig.* 4—Relationship between change in scattering coefficient and energy consumed in loading/deloading cycles (Nordman, Gustafsson and Olofsson)

we have no difficulty deriving qualitative relationships based on common experience, or correlations of trends in the various paper properties with a major change in processing such as amount of beating and refining. The numerous excellent researches on the stress/strain of paper have shown the relationship of load to elongation for variations in the fibre constituents and sheet structure. Empirical relationships such as that of  $Rance^{(35)}$  between folding strength and a combination of tensile load and flexural rigidity are of value in increased knowledge of the factors that affect a complicated mechanical test such as folding endurance.

Only in the case of tensile strength and bursting strength has a direct relationship been established on a reasonably quantitative basis and it is therefore of interest to examine this relationship in some detail. Carson and Worthington<sup>(47)</sup> found that the relationship can be expressed by Pr=2F, where P is the bursting pressure, r is the radius of curvature of the diaphragm at the moment of burst and F is the tensile breaking load in the machinedirection. They reached the conclusion that bursting strength is determined by the machine-direction tensile strength of the paper and that it reveals nothing that could not be determined directly by a tensile measurement. Strachan<sup>(48)</sup> discussed a number of correlative formulae and rejected that of Carson and Worthington and of others as inconsistent with empirical evidence. Strachan claimed that there was a linear relationship between bursting and tensile strengths, provided a formation constant was introduced. The earlier correlations reported were, in general, empirical. Marx<sup>(49)</sup> noted that this problem required closer mathematical analysis than had hitherto been provided. Vollmer<sup>(50)</sup> has provided an analysis of the problem in terms of tensile and stretch and it is interesting to note this development in relation to that of Carson and Worthington.

In addition, Roberts<sup>(51)</sup> and Owen<sup>(52)</sup> stated that tensile strength could be measured in the burst test by using an oblong aperture in place of the circular one. This presumably assumes that the stretch is constant. Harrison and Underhay<sup>(53)</sup> concluded that the low stretch of the machine-direction controls final rupture, but Rance<sup>(33)</sup> showed by experiment that the strongest line must be broken before the diaphragm can actually burst through the paper, even though the weaker has already broken incipiently. Sapp and Gillespie<sup>(54)</sup> showed in commercial papermaking that, if the elongation is sufficiently decreased by tension during drying, it is possible for the paper to lose bursting strength while gaining tensile strength.

Basically, the bubble has been used as a model and, in the simplest physical form, it involves the balancing of forces due to internal pressure and to stress resistance.

Thus  $pa=2\sigma$  (Vollmer)

or PR=2T (Carson and Worthington),

where p, P = internal pressure,

a, R = radius of the sphere at the moment of burst,

 $\sigma$ , T=tensile strength per unit width.

The assumptions inherent in this model are that the paper is isotropic and that the bubble is a portion of a sphere. Since the radius of curvature and the extension are uniquely related, it seems reasonable to express the radius of curvature as a function of the stretch at failure, corresponding to the tensile strength at failure when the bubble bursts.

Thus, according to Carson and Worthington-

$$P = \frac{2T}{R} = \frac{2T}{f(E)}$$

at failure, where E is the percentage elongation.

The function f(E) for R is derived from—

$$E = \frac{200R}{C} \arcsin \frac{C}{2R} - 100$$

where C is the diameter of the orifice.

Carson and Worthington used a graphical method for obtaining R from E. Their conclusion that burst is determined by tensile is true, assuming the concurrent measurement of stretch.

Their conclusion that burst depends on machine-direction tensile only is at variance with Vollmer<sup>(50)</sup> who concluded that burst depends on the average of machine-direction tensile and stretch and the cross-direction tensile and stretch. Presumably, for a given pulp, E is constant, hence P is essentially linear with T. Vollmer uses a geometric device, the arc tan series, to approximate the relationship between radius of curvature and stretch, arriving at-

$$p = 4.9\sigma\sqrt{\epsilon} (1 - 1.5\epsilon)$$

where  $\epsilon$  is the fraction extension and there is, by this means, no need to resort to graphs.

In Fig. 5, YVZ is the profile of the bubble across the diameter YZ of the aperture, WX is the diameter of the sphere WYVZX, of which the bubble YVZ is a part, WX is parallel to the aperture YZ. Vollmer proposed that, for burst, the bubble can be regarded as two cylinders superposed (axially at rightangles) for the case of anisotropic paper.

 $p = \frac{2\sigma}{a}$  for the sphere Since  $p = \frac{\sigma}{a}$  for the cylinder, and

 $p_{\rm cyl} = \frac{1}{2} p_{\rm sphere}$ then





Fig. 5-Relationship among burst, tensile and stretch of paper

	Vollmer	Carson and Worthington
Radius of aperture	r	<i>C</i> /2
Radius of sphere at failure	а	R
Angle subtended by aperture at centre of sphere	2α	
Elongation at failure	E	<i>E%</i>
Internal pressure at failure	р	Р
Tensile strength of paper per unit length, cm	σ	$T_{i}$

He thus uses the average of the two spherical models for MD and CD to give the burst for the anisotropic case, in which  $\sigma_{MD}$ ,  $\epsilon_{MD}$  and  $\sigma_{CD}$ ,  $\epsilon_{CD}$  differ.

Actually, the required relationship between a and  $\epsilon$  can be arrived at somewhat more simply by noting that—

$$\sin \alpha = \frac{\alpha}{1+\epsilon} = \frac{r}{a}$$

where  $2\alpha$  is the angle subtended by the burst aperture at the centre of the bubble and eliminating  $\alpha$  and sin  $\alpha$ . Unfortunately, this is not explicit in  $\alpha$  and

 $\epsilon$ , but recourse to the sine series results in the same approximation reached by Vollmer, alternatively—

$$p = \frac{4 \cdot 9\sigma \epsilon^{1/2}}{r(1+\epsilon)^{3/2}}$$

which avoids one of the two approximations made by Vollmer.

The assumptions made in such a derivation, by either route, are not entirely valid. It is well known that terminal burst and tensile values depend upon the rate of application of the load. It might be possible to contrive a relationship between two parameters, measured under arbitrary conditions, that are each a function of time by manipulation of experimental conditions, but it is apparent that any experimental confirmation of p as some function of  $\sigma$  is somewhat suspect. In addition, the bubble is not spherical. It is a very complex three-dimensional shape, measurable with difficulty and mathematically describable with even greater difficulty. It is clear that the extension of an incremental area of the bubble is maximum at the centre of the surface of the bubble (that is, at the apex) and that the extension at the peripherv of the bubble, tangential to the perimeter of the bubble, is zero. Experiments with aluminium foil reported by Cottrall<sup>(55)</sup> have shown that the rupture commences at the apex-that is, failure when limiting strain is reached. If stretch were uniform, failure would begin at random anywhere on the surface. With anisotropic paper, the meridians vary in contour in every direction and sets of parallels vary non-linearly from side to side of the bubble. The mathematical problem then is the resolution of the shape of the surface of the bubble in three dimensions by integration of the behaviour of the elemental area over the entire surface; this not only at the ultimate failure, but dynamically as the pressure is applied, hence as the strain is occurring. The rate of stretch of each elemental area must be defined and related to the stress/strain of the paper under tensile loading for each corresponding rate, the distribution of the applied stress over the entire surface of the bubble, as well as the pattern of work done and the rate of work absorption. Such an analysis would make it possible to relate the burst test to the fundamental stress/strain time behaviour and make it possible also to describe the point of initial failure and the subsequent path of the failure that is due to the redistribution of stresses. Finally, it might be concluded on this basis that the bursting test would not only be more generally meaningful, but would also yield in a convenient manner the basic relationship between certain fundamental characteristics of the sheet and applied forces.

#### Tearing strength

A PARTICULAR consideration of tearing strength in terms of basic characteristics of paper would seem advisable in this discussion, both because tear is largely negatively correlated with tensile and burst and because tearing stress is the only one that is highly localised in its application.

The work expended in tearing the sheet is the product of the force required to continue the tear and the distance over which the force operates. In this connection, at least two other factors must be included. Splitting of the paper during the tear will obviously increase the amount of work required in view of the larger area of rupture. In addition, bending of the sheet is involved and it would be expected therefore that the work expended would increase with sheet rigidity.

In general, it is known that tear varies inversely with the density of the sheet beyond an initial rise with an early portion of the density increase. When the density increase has been brought about by beating, it might be suspected that the tear decreases at least partially as a result of the accompanying fibre shortening. It is recognised that tearing strength is strongly dependent on fibre length. Since wet pressing alone results in a decrease in tear, however, it is apparent that increased density through the accompanying increased interfibre bonding lowers the tear strength. This is further shown by the effect of the addition of extraneous bonding or cementing agents in lowering the tear of sheets made from unbeaten pulps.

An interesting theory<sup>(19)</sup> has been presented to explain these wellrecognised facts. In this analysis, it is suggested that the average work expended in fibre rupture is relatively small in comparison with that required to pull intact fibres out of the web, although the force required for the latter is somewhat less. The average tearing force required is then given by the sum of the two types of work divided by twice the length of the line of tear. Thus, the tearing strength will be governed by the relative numbers of fibres ruptured and fibres pulled out intact. On this basis, the initial increase in tearing strength in the early stage of beating is explained by a tighter enmeshment of the fibres, in which very few fibres are ruptured, but the frictional drag work of pulling out intact fibres has been increased. As beating continues, more and more fibres fail in rupture and fewer are pulled out intact, presumably as a result of continuously increasing interfibre bonding. Since fibre rupture involves much less work, decreased tearing strength results. The same would hold true as a result of the addition of an extraneous bonding agent.

This general analysis of tearing strength would appear to offer a satisfactory concept of the relationship between tearing strength, interfibre bonding and sheet strength. The experimental finding of Van den Akker, Lathrop, Voelker and Dearth<sup>(28)</sup> that tear involves a preponderance of fibre rupture rather than fibre pull-out is very interesting in this connection. Increased fibre length would obviously result in increased work required for fibre pull-out, hence in increased tearing strength. It might be expected that this relationship would diminish in importance as bonding saturation is approached and investigation of this factor would be of considerable interest.

The above concept might be regarded also from the point of view of the degree of restriction or concentration of the tearing stress over a decreased area of paper—or the involvement of fewer fibres—with increased interfibre bonding. It is apparent that diffusion of this stress over a larger number of fibres would result in increased work of tear, hence a higher tearing strength. Mason<sup>(56)</sup> has noted that high tearing strength is associated with high creep, which tends to distribute the load over a wider area. Thus, the loss of tear on moistening a sheet is much less than the loss in tensile. Steenberg<sup>(57)</sup> noted that microcreping in the paper structure must result in increased tearing strength. In this relationship, the degree of strain produced in papermaking operations is of considerable importance. Steenberg<sup>(57)</sup> showed that, if a machine-made paper is strained in the cross-direction, an appreciable loss in tearing strength ensues. In the machine-direction, however, where reasonably tight draws were employed during manufacture, little difference was brought about by prestraining. Thus, the elongation available in the cross-direction, whenever drying is carried out with relatively little restriction, is an important factor in tearing strength.

Wahlberg<sup>(58)</sup> has presented a quantitative confirmation of this relationship between tearing strength and extensibility. Machine-made paper taken from near the edge (where cross-direction elongation is high) was strained to varying percentages of the breaking load. Then the tearing strength and strain at break were measured very quickly (within 30 sec) on these test samples. It was found that the tearing strength and strain at break both decreased with increased preloading. He suggests that appreciable force is required to straighten out the microcreping. The decrease in tear becomes sharper at higher degrees of prestraining, denoting presumably the greater removal of creep at the higher levels of preloading. In the same sense, it was shown also by Wahlberg that the greater the tension under which paper is dried, the lower is the tearing strength. Higher tension results in less contraction, hence less microcreping, since the web is dried in a stretched condition. The degree of microcreping should vary across the sheet depending upon drying conditions. Measurements across the reel on machine- and cross-direction tearing strength, together with extensibility at the same locations, showed a good correlation between tear and stretch.

Giertz and Helle<sup>(59)</sup> have measured the tearing strength, after various amounts of beating, of sheets dried completely without restraint. They showed that after an initial decrease the tearing strength remained constant over a wide range of beating. This constitutes a striking illustration of the importance of microcreping on tearing strength. Arlov and Ivarsson<sup>(60)</sup> have shown a close relationship between tearing strength and strain at break. Higgins<sup>(61)</sup> was not able to confirm the above correlation between tear and stretch, particularly in the highly beaten region for Eucalypt kraft paper of the same structural pattern.

Jones and Gallay<sup>(62)</sup> showed that tearing strength per unit weight of paper varied with the basis weight. They concluded that the increased rigidity of the heavier weight was not the explanation for the increased tearing strength obtained, but rather that the increased amount of splitting constituted the chief factor involved. Wahlberg<sup>(63)</sup> obtained a linear relationship between tearing strength and stiffness, but concluded also that increased stiffness was accompanied by increased splitting, thus bringing his work into general agreement with that of Jones and Gallay.

In an examination of the papermaking qualities of fifteen species of hardwoods, Tamolang and Wangaard<sup>(64)</sup> conclude from multiple correlations that tearing strength of paper made from beaten pulp is primarily dependent on the strength of the fibre, with fibre length also of considerable importance. In papers made from unbeaten pulps, they conclude that, in addition to fibre length, increased lumen width and decreased cell wall thickness lead to increased tearing strength. This must be interpreted as showing a positive correlation between fibre flexibility and tearing strength for unbeaten pulps. This is in general agreement with the increase in tearing strength during the first stage of beating, referred to earlier, since the effect of the latter would be to increase the fibre flexibility. Only at a subsequent stage does a marked degree of bonding bring about the well-known adverse effect on tearing strength. It is quite generally noted also in mill-scale operations that this initial increase in tear is obtained with sulphate woodpulp, but not with sulphite pulp. This is in all likelihood related to the greater density shown by unbeaten sulphite pulp, resulting in turn from certain considerations of fibre dimensions and fibre damage in this pulp.

Although tearing strength like all other properties of paper is dependent on fibre properties, interfibre bonding and fibre arrangement in the paper, it would appear in the normal range of papers that fibre properties are of particular importance to tearing strength. When an appreciable amount of interfibre bonding has been introduced, failure occurs mainly through fibre rupture, hence the strength of the fibre probably constitutes the outstanding basic characteristic required. For any given fibre strength, however, tearing strength will be greatly altered by the degree of diffusion of the applied force. In the extreme case of a thoroughly bonded sheet with fibres locked into position in such a manner that movement within the web is obviated, the tearing force can be visualised as shearing off each fibre in turn in the line of



tear, with the force highly concentrated over this narrow path. Whenever slight alignments or dislocations between points of bonding are possible, however, the applied force is taken up over a much greater area and the work of tear is substantially increased. Consequently, the factors involved in restrictions during drying (for example, the machine-direction tension) will have an important bearing on the tearing strength. The length of the fibre is also of considerable importance in assisting in this ability to diffuse the applied force. The concept of microcreping as used by Steenberg (Fig. 6) is a useful one for a schematic representation of the type of structure valuable in this regard. It is interesting to note, in this connection, that the recently developed commercial papers that are deliberately shrunk during drying to produce an extensive amount of such microcreping show greatly improved tearing strength.

#### Handle, softness, hardness and compressibility

IN some instances, commercial evaluation of paper is made purely on the basis of subjective judgment and this, in fact, is of outstanding importance in the acceptance or rejection of paper in the first instance, guite aside from functionality during actual usage. The so-called handle of paper (analogous to the 'hand' of a textile fabric) has steadily become of increasing importance in recent years in fine papers for a variety of printing and writing purposes. In judging such papers, the evaluator feels the sheet, rattles it, frequently flexes the sheet into compound curvatures with accompanying relatively sharp folds at angles to one another in the sheet. Brecht<sup>(65)</sup> has given a review of the literature on the subject and an account of his own work in attempting to relate handle, as judged by panels of expert evaluators, to individual physical properties of the sheets. Brecht came to the general conclusion that the handle of paper is a function of the square of the flexural stiffness divided by the thickness of the sheet for any given basis weight. There is no doubt that the handle of paper improves with flexural rigidity for any given thickness. The formula of Brecht also states, however, that the handle improves with less thickness for any given rigidity and this is interpreted to show the importance of hardness as a factor in handle. This is undoubtedly broadly true, for example, if we compare a distinctly soft paper with a hard paper, both of the same weight. Here, the thickness of the soft paper would have to be much greater than that of the hard paper in order to attain the same flexural stiffness. If the difference in hardness is great, there is no doubt that the thinner, harder paper would be chosen by the appraiser as being superior. Nevertheless, some observers stress bulk (inverse of density) in judging reasonably similar papers and this emphasis on thickness would suggest that the relationship chosen by Brecht would not satisfy such observers. Handle of paper has no distinct definition, is subjective and is therefore open to individual interpretation. This contribution of Brecht with its numerical expression of a complex subjective property of paper constitutes an important advance in our knowledge of the subject.

Here also, it is noted that the density of the sheet is of primary importance. For any given basis weight, a lower density (greater thickness) gives a feeling of greater body and therefore of better value. At the same time, this lower density contributes to a greater flexural rigidity, which varies as the square of the thickness for any given weight. With too low a density, however, the accompanying lack of bonding will decrease the rigidity both in flexure and in compression. There is, thus, a compromise among basic properties and this compromise will be materially affected by the flexural rigidity of the fibres of which the sheet is composed.

Another property in part subjective in nature is the softness of paper. Lyne<sup>(66)</sup> has analysed factors entering into softness and has included density, rigidity, compressibility and surface smoothness. Flexibility is particularly important in this regard, again a low density is required, adding also to the compressibility. Suitable steps in manufacture are taken in these directions, particularly in the avoidance of beating and further decrease in bonding is achieved frequently by dry creping. With facial and sanitary tissues particularly, also with towelling, a softness of surface is required as well. This is akin to the velvety feel given by a fibre floc on a surface and constitutes a particular feature of softness that is highly subjective and difficult to measure by test.

Compressibility of paper should be regarded as one of the factors entering into the general stress/strain properties of paper in the thickness direction. This is a field that has apparently been virtually entirely neglected, despite the likelihood that such investigation would enhance greatly our knowledge of the structure of paper. This is in addition to the importance of such information in various functional applications of which printing is an obvious example. Here again, it is likely that the basic factors of sheet density and flexural modulus and resilience in flexure of the fibres are governing factors. Hardness of paper is obviously closely related to density.

#### General discussion

In actual practice of the utilisation of paper, a great variety of demands on the properties of paper are encountered for specific applications. It should be emphasised at the outset that the actual properties required of the paper are in many instances only partially understood. Frequently, especially on some of the machinery used for converting paper to its final form, little or no attempt has been made to analyse the forces applied to the paper, hence the properties that must be built into the paper for these specific purposes. It is no exaggeration to say that, in many mills, the traditional measurements of

35—F.S.P. i

bursting and tearing strengths of the paper still serve to characterise a variety of papers with little regard to the actual properties demanded. It is apparent from qualitative correlations such as have been described in an earlier section that certain interpretations of burst and tear data can be inferred on the basis of some other properties, but the inherent dangers of such a procedure are obvious and such an interpretation is in any event bound to be woefully incomplete for a suitable understanding of the situation in most instances.

Failure tests following the various kinds of applied stresses bear little relation, except in a rather remote manner based on loose correlations, to many applications. At times, they can be actually misleading, as in well-known instances of using a tensile load at failure in place of work absorption in the stress/strain course of the test. The term *strength* is frequently used without qualification and it is apparent from everyday experience that the type of strength required must be defined—for example, tensile or tear—for the word to have real meaning. Many important properties of paper such as softness. hardness, brittleness and pliability are governed by the ease of deformation of the sheet under comparatively mild stresses far removed from that required to bring about failure. The prerupture behaviour of the paper in stress/strain and consequent information on elasticity, permanent set, secondary creep, stress relaxation, frozen-in stresses and strains and the like are frequently of far greater value than levels obtained in arbitrary failure tests. This is true not only for purposes of evaluation of the paper for particular applications, but also from the point of view of obtaining information on the structure of the paper and on the nature of the component fibres.

In preceding sections of this discussion, various properties of paper have been related within the framework of existing knowledge to a group of basic characteristics of the sheet. These represent the properties of the component fibres, their arrangement in the sheet and the extent to which they are bonded together. The basic sheet characteristics in question-namely, density, fibre dimensions, fibre strength, fibre orientation and interfibre bonding-are fixed by fibre properties and processing variables on the papermachine. It should be emphasised that the fibre properties referred to here are those that obtain at the head box of the papermachine. They reflect, therefore, all the changes in fibre properties brought about in all the previous mechanical and chemical processing applied, including particularly chipping, pulping, bleaching and stock preparation. The alterations in fibre properties that result are of outstanding importance in their reflection on the structure and properties of the sheet produced. On the papermachine, certain processing variables are available to the papermaker to adjust properties within limits to conform to specifications. The relative importance of fibre properties and papermaking

techniques toward the production of high quality paper is still frequently the subject of some discussion, particularly in integrated mills. The pulpmaker may feel that his good pulp is subjected to something less than optimum treatment on the papermachine, while the papermaker may be under the impression that his best efforts cannot cope with the lack of quality of the pulp that is provided to him. It is of interest to note that some extensive experimentation was carried out by this writer several years ago in this connection. Pulp properties, papermachine variables and paper properties were measured over a considerable period, involving several grades of paper. Multiple correlations of the data produced left no doubt about the overwhelming importance of variation in pulp properties, even with the normal swings in papermaking practice. Formation can, within limits, be adjusted on the machine; yet here also. pulp properties exert a considerable effect. The same is true to a lesser degree for fibre orientation. Some adjustment in sheet density can be made by variation in wet pressing, but the fibre properties involved are far more important in this regard. Tension applied during drying is of considerable significance, regardless of fibre properties. The effect of calendering depends largely on fibre properties. It is not intended to infer by the foregoing that processing variables on the papermachine are of little consequence; it is intended rather to point out that they are of less significance than fibre characteristics for paper properties and are, as a matter of fact, to a great extent dependent on these fibre characteristics.

In earlier sections of this discussion, some attempt has been made at a separation of the effects on paper properties and structure, of individual fibre characteristics-fibre dimensions, fibre strength and wet flexibility of fibres. Nevertheless, it is apparent that these characteristics are to a great extent interdependent, particularly to the action of beating. In the flexibilising of the fibre (and the exposure of active polar groupings on the fibre wall), the severe mechanical action of beating brings about a degradation in both length and strength of the fibre. The extent of this degradation for any given type and amount of beating depends on both the morphology and the history of the fibre. Springwood fibres undergo breakage and other forms of damage much more readily than the thicker-walled summerwood fibres. The sufficient development of zones of damage along the length of what was considered a long fibre would be expected to result actually in an approach to the equivalent of a number of very short fibres, even when complete breakage has not actually occurred. This is strikingly shown in the work of Forgacs<sup>(6)</sup> and of Forgacs, Robertson and Mason.<sup>(7)</sup> It is apparent that the fibre length to be considered is not that of the original fibre (or even what may be roughly measured by a screen classification or other type of measurement on the beaten pulp), but

rather that of the 'effective length' of the fibre in the web. Thus, beaten sulphate and sulphite pulps made from the same spruce chips cannot be considered as having the same fibre length.

It is difficult to differentiate this effect of beating on fibre length from the accompanying effect on fibre strength. Fibre strength depends (aside from broad differences in form such as with softwoods and hardwoods) on the intimate internal structure of the fibre, the orientation of substructures in the fibre, the nature of the intrafibre bonding and on flaws in these intrafibre characteristics. The strength of the fibre in shear or even in tensile normal to the fibre axis are of primary importance to the properties of the sheet. The tensile strength of the fibre in the axial direction must be considered as only one aspect of fibre strength. In this discussion, the term fibre strength is used in the sense of involving the integrity of the fibre in general toward stresses that may be applied to the paper or, in fact, that may be applied also during paper manufacture (for example, in drying on the papermachine). The term fibre strength applies, therefore, to resistance to rupture of a portion of a wall, as well as to breakage through the fibre in tension or in flexure.

The relationship of fibre strength with interfibre bonding seems worthy of consideration. It was suggested by Gallay<sup>(29)</sup> that failure under stress would be likely to occur within the fibre adjacent to the interfibre bonded area rather than in the bonding zone, particularly if fibres had been subjected to marked damage in processing. The work of Jayme and Hunger<sup>(67)</sup> appears to give considerable supporting evidence to this concept. McDonnell and May<sup>(68)</sup> have noted the type of damage brought about by hypochlorite treatment, involving wrinkling, cracking and severing of outer wall structures in the fibre, which would be expected to lead to intrafibre failure rather than to interfibre bond failure on stressing. Thus, the strength potential of interfibre bonding can be realised only to the extent that the fibre strength remains at a reasonable level. The work of Leopold and McIntosh,<sup>(69)</sup> relating the tensile strength of the fibre to xylan content, is of particular interest in this connection, suggesting that hemicelluloses are important not only for interfibre bonding, but also for fibre strength itself.

Before forming the web, fibre flexibility and plasticity in the wet state must be attained to varying degrees depending on the properties desired in the final web. Density of the sheet, consequently interfibre bonding (assuming proper preparation of the fibre wall), will depend on this wet flexibility. The ensuing properties of the finished paper will depend, however, not only on density and extent of interfibre bonding, but also on the fibre strength and length that the fibres possess in the web. Thus, for example, all strength properties of paper will increase with fibre strength and fibre length, including tensile, burst, tear, fold and rigidity. The case of tearing strength is particularly interesting in this regard. Tearing strength is generally regarded (and rightly so) as opposed to tensile and bursting strengths. This results from the fact that increasing degree of interfibre bonding localises the tearing force and lowers tearing strength, whereas increased interfibre bonding assists in resistance to failure in tension and burst (up to a certain maximum). Nevertheless, at any given state of interfibre bonding, tearing strength varies with fibre strength and length as do tensile and burst; as a matter of fact, the dependence is probably greater in the case of tear. Fibre strength and fibre length, particularly in the sense in which they are used here, are thus fundamental fibre characteristics required for all strength properties. Permeability properties also are concerned, since the formation of debris (denoting to a large extent lack of fibre strength) has a serious effect not only on the drainage characteristics of the pulp suspension during processing, but also on the flow of fluids through paper. Opacity is generally related only to density and related interfibre bonding. For any given sheet density, however, opacity also is materially affected by fibre length and indirectly, therefore, to fibre strength. It was earlier noted that sheet density is dependent primarily on the wet flexibility of the fibres, assuming preparation of the fibre wall for interfibre bonding. It is apparent, however, that the segmented type of bending shown by Forgacs, Robertson and Mason<sup>(7)</sup> will greatly affect this factor of gross wet flexibility, even when relatively little flexibility has actually been attained in the fibres within the segments themselves. The relationship normally expected between high density and high tensile and bursting strengths may be markedly disturbed by this factor. It is frequently found that a pulp that has been seriously damaged (for example, by excess hypochlorite in bleaching) shows a relatively high density even before beating, as well as after beating, but the tensile and bursting strengths are comparatively low. This attainment of a higher density through faults along the fibre such as hinged joints resulting from nodes or the cracking of embrittled fibres would be expected to have an important effect on the stress/strain properties of the sheet in the thickness dimension. Fibres having these faults would retain compressive strains to a much greater extent, thus the resilience following calendering action would be reduced. The differences among various pulps in this regard can be very marked. This is directly related to the compromise between smoothness of surface and bulk of the sheet, for which, as frequently applies particularly in printing and writing papers, a maximum retention of bulk with attainment of the desired smoothness is important. There is thus suggested a relationship also between these properties of paper and the strength and effective length of the fibres.

In the foregoing, the dependence of various basic characteristics on the effective length and strength of the fibre has been discussed. It is obviously impracticable to compare directly a short, thick-walled fibre with a long fibre of quite different axis ratio. For any given species of wood, however, the effective length will depend on the strength of the fibre, since both shortening of the fibre and flaws along the fibre will depend on the resistance of the fibre to the action of beating.

In a consideration of the properties of paper and their interrelationship, therefore, it is suggested that fibre strength in the broad sense as it is used in this discussion is of outstanding basic importance.

Particular emphasis is placed on this factor, since the impression is gained from a study of the literature on various aspects of this matter that insufficient attention has been paid to fibre strength in explanations and discussions of observations made. Fibre strength for any given species of wood is fixed by the extent of degradation suffered by the fibre from the tree to the reel of the papermachine and each processing step will react in this respect to what has gone on before. Thus, chipper damage, weakening of the fibre through cooking and bleaching and, finally, degradation through beating are all interrelated.

In general, in this discussion, an attempt has been made to examine a sheet of paper from a point of view that would allow for the derivation of all the common properties from more basic considerations. It is considered that this is the most logical viewpoint to be taken in an examination of the interdependence of the properties of paper. In addition, it would appear that such a viewpoint has the merit of unification or 'wholeness' of paper properties, which is deemed to be preferable to a characterisation by a number of separate categories of properties difficult to relate directly. Finally, it has been suggested that these basic characteristics are themselves interrelated and that, within ranges of fibre forms, the maintenance of integral fibre strength in the broader sense is a predominant factor in governing the properties of paper.

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### **Transcription of Discussion**

## DISCUSSION

PROF. J. D'A. CLARK: Gallay is perhaps one of the most able exponents of what someone—maybe myself—has very impolitely called the 'boiled macaroni theory'. Because of this particular concept of the bonding of fibres, we may stray from some of the more fundamental relationships between fibres and the resulting paper. For example, Gallay refers to the work in 1934 of Ainsworth Harrison, whose published photographs show no fibrils on a beaten fibre. In 1941 or 1942, I published photographs of fibrils present on slightly beaten fibres after silvering, which was confirmed by electron micrographs of the same fibres (among the first micrographs of fibres to be published). We did not make it clear at the time that there are no fibrils on the primary wall, if the fibre is once dried. On the other hand, when the fibre remains wet after pulping, there are these almost invisible microfibrils on the primary wall.

Many of the relationships between fibre properties and paper cannot be explained without postulation that these fibrils are actually there. Unless silvered, these microfibrils can be seen in an optical microscope only with the greatest of difficulty and they are collapsed when a specimen is dried out for electron microscopy.

It may be well to refer further to the effect of fibre length on the properties of paper. This last year, I have been confirming my work mentioned by Gallay about the influence of the weighted average fibre length by weight on various properties and their relationships with paper having a moderate amount of bonding. The relationships are only semi-quantitative, since the curves are sigmoidal, but the main slopes are in conformity with my quoted earlier published findings. I found, too, that, if the fibres are the equivalent of well-beaten stock with more adhesiveness, the slopes of the curves decrease. For example, the relationship of the normal folding test to weight average length (L) varies as its cube for very well bonded paper. The cubic relationship for fold drops down to the straight L factor with very well bonded fibres. The bursting factor, which is proportional to the first power of the length, drops to about half. I might add that the reason for different exponents for the length factors for burst and tensile strength is that burst comprises two factorsstretch and tensile strength. Each one of them is related to the half power of fibre length, so the two together give the first power relationship for burst alone.

#### Interdependence of properties

It might have been well to have referred also to the work of Boyd Campbell in connection with bursting strength. He indicated that even with parallel cotton threads, in bursting the structure, a perfect sphere was obtained; consequently, the burst is strictly a relationship of the strength and the stretch in the machine-direction. Some of Doughty's quoted results on fibre length are vitiated by the fact that, because of the normal length/ coarseness relationship for wood fibres, some of his findings were determined by the fibres being finer rather than shorter. Moreover, as Gallay has mentioned several times in his paper, my experience in no way indicates that sheet density increases with fibre length. Such an effect is very slight and, if anything, I have observed only a fall in density with increased length.

Finally, I consider that some phenomena like that pointed out by Gallay in which a sheet made of fibres that have been dried by the displacement of liquids increases in strength after keeping in a moist atmosphere can be explained satisfactorily only by the fact that the microfibrils are able to condense moisture from a moist atmosphere by virtue of their fineness, hence it increases the number of those that can subsequently bond together.

MR. N. C. UNDERWOOD: We were given a warning about not deriving interrelationships among tests, but do these not have value as an internal check on the results from several instruments?

Where does the energy to rupture fit into the picture?

DR. W. GALLAY: Of course, I did not intend to convey that this list of sheet properties is by any means complete. Many others could be taken into account. I will answer the question in the most general terms by saying that I would include energy of rupture among those properties that depend so strongly on the cumulative history of the fibres.

**PROF.** G. JAYME: It is a very large field that Gallay has been opening and I have the feeling that the time available is insufficient, but I intend to show some slides tomorrow about the interrelationships of fibre properties and paper strength, taken from another paper.<sup>1</sup>

DR. GALLAY: I mentioned quickly in passing that we require some form of measurement for what I have termed *effective strength* (I have described in considerable detail in the written paper the meaning I attach to this term). In this connection, I have been much impressed with the experiments performed by Jayme and by Stone, involving the differentiation of fibres by resistance to

<sup>1</sup> Jayme, G., Das Papier, 1961, 15 (10a), 581-600

#### Discussion

swelling and solution. This might constitute one avenue of approach toward such an evaluation.

The statement has frequently been made in the literature to the effect that density is the dominant sheet property. You will note that I have placed density somewhat subservient to strength for reasons given in my paper. It is everyday experience that we can in fact obtain pulps high in density and with strengths varying from high to low.

MR. J. D. PEEL: Has Prof. Giertz examined sheets made from hardwood pulp, dried with and without restraint, for their tearing characteristics (in the way he described the examination of sheets made from softwood pulp)?

PROF. H. W. GIERTZ: No, we have not done so.

MR. H. G. HIGGINS: I would like to answer that question, because we have done so with hardwood pulps and been unable, as Gallay pointed out, to confirm the relationship between extensibility and tear factor. There may be a small effect, but nothing like as much as that obtained with long fibres. Extensibility is not nearly as conservative a property of the material as tear factor. We can change the extensibility in various other ways apart from inducing microcreping—in ways that do not influence tear or may affect it in the reverse direction. Just why there is this difference between hardwoods and softwoods, I am not quite clear.

I would add that I fully agree with Giertz' reason for the decline in tear on beating being due not so much to fibre shortening as to the improved fibre bonding.

MR. T. TROMP: With 'papers' made from synthetic fibres of known length, the degree of artificial bonding is increased, the tensile and bursting strengths increase while tearing strength falls. When longer fibres are used, the tearing strength increases with the same amount of bonding. This illustrates that the effect of fibre length and bonding is not in any way connected with the effects of beating.