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CONSEQUENCES OF THE LAYERED STRUCTURE OF PAPER

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Synopsis The layered structure of paper is a necessary consequence of its method of manufacture by deposition of fibres from a low concentration suspension in water or air. It results in an extreme anisotropy of the in-plane and out-of-plane properties. Reported and estimated values of compressibility, strength, permeability of fluids and light transmission, in the two directions, are compared, although such comparisons are often only speculative in the absence of direct measurements of the properties of non-layered papers.

The total result of the layered arrangement of fibres is a material that is relatively dense and smooth, stiff, strong in tension, but liable to crease and delaminate. Such a combination of properties determines its performance in the four main areas of application—

1. For printing, it offers a compact, smooth surface, but poor processing strength and opacity.
2. For packaging, good puncture resistance and easy creasability of board, but poor folding endurance.
3. For hygienic and disposable products, good processing strength, but poor softness and absorbency.
4. For barrier and filter media, good strength, but a dense packing.

Attempts are discussed that are aimed at overcoming these limitations of the layered structure.

Other sheet materials, such as felt and leather, possess a non-layered structure that strongly affects their performance. The possibilities are discussed of extending the areas of application of paper and of paper-like materials through making its structure non-layered.

Introduction

PAPER and paper-like sheet materials possess a unique type of fibrous structure: their component fibres, although much longer than the thickness of the sheet, are each disposed at different depths with little penetration in

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the Z-direction (that is, normally to the plane of the sheet). The fact appears obvious from examining microphotographs of cross-sections of paper[1] and may be estimated quantitatively.[2] In contrast, some natural sheet materials such as leather contain fibres and bundles of fibres, which extend through the whole thickness—an excellent example may be seen in Kerner's work.[61] Similar effects occur in woollen felts and, of course, in woven materials.

The layered structure of paper results from its method of formation, by filtration from a dilute suspension;[2] the consequent effects on the physical and use properties form the subject of this review. In the absence of published data on the properties of paper with a non-layered structure—the author has failed to find a report of such an article—any statements on the effects of such a structure must necessarily consist of theoretical predictions; of indirect comparisons between dissimilar materials; and of pure speculation. All these three approaches are used in the following text.

The first part reviews the available information on the effects of the layered structure of paper on four main groups of physical properties: thickness and compressibility; strength; porosity; the optical properties. The consequences for the use properties of paper are then briefly discussed.

It appears that they are often of only marginal importance for most papers in their conventional applications. With the extension of the papermaking and similar techniques to the newer fields of non-woven textile replacements and of fibre reinforcement of plastics, however, the importance of the layered structure becomes greater. The second part of the paper deals with the effects of the non-layered structure of some materials other than paper, also with the published accounts of the papermaker's attempts to produce such structures.

Effects of the layered structure of paper and board on their physical properties

BOTH paper and board may be considered as dispersions of cellulose in air. When the elementary particles are roughly granular in shape, the theory of Kerner[3] applies with considerable accuracy, as demonstrated by Baxter,[4] but this treatment need not extend to predicting the behaviour of fibrous-shaped particles and especially the effects of their different arrangements. In contrast, the theories of fibre reinforcement of solid matrices give some insight also into the behaviour of paper, even though the continuous matrix here is not solid. The theories of paper structure usually simply assume that it is layered.[5]

Thickness and compressibility

These two concepts are interchangeable, since the measured value of thickness depends on the pressure of the anvil. The power law of compression, which applies to a very large number of expanded materials and over large ranges of pressures[6] is valid both for the three-dimensional structure of woollen felt and for the layered structure of paper and so offers no means of distinguishing the two types of structure. Intuitively, one would expect the latter to be the more compressible.

Measuring the compression response of unbonded mats of nylon fibres, Elias[7] concluded that the presence of some 4 per cent of the fibres oriented in the Z-direction largely determines the compression response, making the mat less compressible; however, he produced no quantitative estimate of the effect.

Working with papers formed under vacuum, Chang[8] found that samples made under conditions simulating those on a papermachine were some 20 per cent bulkier than laboratory samples made from the same stock, especially when measuring the thickness at very low pressures. He attributes the difference to the greater degree of Z-direction orientation of the machine-made papers. Work at the Swedish Forest Products Research Laboratory in Stockholm on forming from suspensions of very high consistency confirms that such papers are bulkier and may be compressed to a high dryness without losing bulk.[9]

I have failed to locate a published reference to a direct comparison of the thicknesses of paper sheets formed in one step or in layers—for instance, the main reasons for producing multi-layered board are not physical effects, but simply the economics of production.[10, 11] Such comparisons are further complicated by the well known difference in the apparent density of single sheets of paper and of stacks of such sheets. It is generally accepted[12–15] that the difference results from the more open structure of the surface layers, which intermesh during the stacking. It disappears if the thickness of the single sheet is measured by a method that allows for the irregularities of the surface (for example, microscopically) or by measuring penetration. Hsu[16] demonstrates that the distribution of such irregularities is log-normal, as would be expected from a layered structure.

Another curious aspect of the measurement of thickness of paper is the apparent negative Poisson ratio: when extended in plane, most papers increase in thickness.[17] Öhrn[18] demonstrates that the effect is small (some 10 per cent), reversible and due to the layered structure of paper. It is much larger and more obvious in the case of non-woven materials, where it contrasts with the behaviour of woven cloth.[19] It is difficult to visualise a practical consequence of this curious phenomenon, although it may be of some relevance to the performance of paper-like structures, when these are used as fibre reinforcement of a plastic matrix.

To summarise, the effect of the layered structure of paper is to make it a little more compressible, especially at low densities. There are indications that some conditions of formation of the web result in an appreciable degree of Z-direction orientation, which may give an increase in bulk of up to 20 per cent, but these indications are, as yet, uncertain and rare.

Strength

It is an observation so obvious as often to escape notice that the bonds between fibres occur only in the horizontal plane of the sheet (for example, see Page & Tydeman's work[20]). Thus, each bond failure is a microscopic delamination and the failure of the sample itself merely the sum of such delaminations. If the structure of paper included an appreciable proportion of fibres spanning the different layers through the Z-direction, then one would expect to observe the breaking of such fibres as part of the failure of the sample. In fact, Helle[21] observes only bond failures throughout the extent of the sample and fibre breaks only in the immediate vicinity of the zone of failure. It appears that the resistance of paper to failure is largely determined by its layered structure; directly so for the Z-direction strength (which is, indeed, often used as a measure of bond strength) and indirectly for the in-plane strength.

It may be worth noting that the strength anisotropy of paper is, of itself, no proof of its structure being layered, since it could result merely from the nearly horizontal orientation of fibres (fibre length being much greater than the thickness of the sheet).

Z-direction strength

Brecht & Blikstad[22] find the tensile strength of paper in the Z-direction to be only 1/30 to 1/100 of that in plane.

Clark[23] observes that some multi-planar laboratory sheets are particularly difficult to split into layers and attributes the difference to a degree of Z-direction fibre orientation. Finger & Majewski[24] observe preferential splitting of machine-made paper in different directions and explain it as due to the presence of discrete layers, each with slightly different Z-direction orientation. Wink & van Eperen[25] correlate measurements of Z-direction ultimate tensile strength with the relative bonded area, as measured by optical scattering: the ratio is independent of density. Predictably, the presence of fines has a larger effect on the Z-direction strength than on the in-plane strength. The earlier estimates by Brecht & Blikstad of the relative magnitudes of the two strengths are largely confirmed.

Clearly, the Z-direction strength of paper reflects its layered structure. One may expect a large effect to result from any small degree of fibre orientation in this direction, but no such findings have yet been reliably reported.

In-plane strength

Again, a comparison with the theories of fibre reinforcement of continuous matrices is instructive. For instance, Nielsen & Chen[26] confirm that the rule of mixtures applies to calculating the strengths and moduli of materials reinforced with disoriented fibres, if the effects of orientation are allowed for by simple integration. Cook[27] finds that a small degree of disorientation out-of-plane of fibres in a laminated fibre-reinforced material has little effect on the in-plane modulus. One would, therefore, expect that a slight disorientation of fibres, such as would result in a non-layered structure of thin paper, would be of little consequence compared with any effects of interweaving or locking together of the fibres.

In fact, no such effects have been reported. Kallmes *et al.*[5] find the stress/strain diagrams of laboratory papers made in single thicknesses or multi-planar virtually identical. The theory of the strength of paper by Page[28] largely confirmed by Burgess[29] implies that fibres penetrate only through a very small proportion of the paper bulk, with most papers of practicable grammages having an essentially layered structure. An interesting theory by Hopkins & Ranger[30] describes the tensile failure of paper as delamination of bonds by a peeling action under a compressive stress. Öhrn,[18] although criticising this theory, agrees with the concept of delamination of a layered structure.

One may justifiably expect appreciable increases in the in-plane strength of paper to result from interweaving of fibres and a departure from the layered structure. In fact, Wahren[9] reports the opposite finding for paper made at a very high consistency, which could be expected to have an appreciable degree of Z-direction orientation—its tensile and bursting strengths are distinctly lower.

Tearing strength

Again, one may expect greater tearing strength from a non-layered structure, if only as a reflection of increased Z-direction strength. Wahren's findings[9] confirm this expectation.

Porous structure

Corte & Lloyd[31] demonstrate that the observed log-normal distribution of pore sizes may be derived from a layered model of the structure of paper. Hsu[16] extends this derivation to the structure of surface pores. Neither theory, however, makes it possible to predict the effect of a non-layered structure on the distribution of pore sizes. Some such prediction may be made from considering the pore tortuosity factor, as defined by Biffer &

Mason.[32] Using such methods, Stamm[33] finds that the average length of pores in paper is some 1·5–2·5 its thickness, in agreement with the layered model. An appreciable degree of Z-direction orientation would be expected to reduce the pore tortuosity factor, but only slightly.

Permeability to fluids

Back[34] finds the value of the Z-direction permeability of paper and board to water only a few per cent of that in-plane; for fibreboard, this ratio may reach some 30 per cent. Didriksson & Back[35] report that pressing fibreboards reduces the pore size anisotropy to a minimum at a density of 0·5–0·8 g/cm^3; further pressing of machine-made boards sharply increases the anisotropy again. This is not so with laboratory-made boards, suggesting a different structure.

Kyan[36] rejects the Kozeny-Carman 'channel' model of liquid permeability in favour of the 'drag' model, mainly because he finds the resistance to fluid flow unexpectedly high for the high values of porosity of most fibrous mats. The latter suggest the existence of a large proportion of dead spaces such as would result from a layered structure (in contrast to Stamm[33]). Bliessner[37] finds that the mean hydraulic radius of pores decreases with increasing grammage up to some 100 g/m^2, presumably because of increasing branching out of pores. Peterson[38] concludes from measurements on very layered beds of nylon fibres that the in-plane permeability is some 30 per cent greater, as predicted by theory.

It appears that the pronounced anisotropy of the pore size distribution and of the permeability of paper to fluids simply reflects the preferential orientation of fibres in the plane of the sheet. The effect of the layered structure is probably marginal, resulting in a slight decrease in the permeability in the Z-direction. The latter is sufficient to make a distinction between the 'channel' and the 'drag' theories of the resistance to the flow of fluids, but is hardly significant from the user's point of view.

Optical properties

The author has failed to uncover any published reference on the effect of the layered structure of paper on its optical properties. Any such effect is likely to be indirect, resulting from differences in bulk and the degree of bonding.

Effects of the layered structure of paper on its usage properties

Paper and board are used in so many and such different applications that it becomes impossible to discuss each in turn. From the considerations above of the effects on the main physical properties, however, a general picture emerges as follows.

Because of their layered structure, paper and board are sheet materials that are marginally denser and more compressible than would be the case if the fibres were oriented in the Z-direction to an appreciable extent. They are also much weaker in the Z-direction for the same reason; probably also weaker in their in-plane tensile and tearing strengths. Their permeability is marginally less at any given density; no indication has been found of how the layered structure affects the optical properties and surface smoothness.

This complex of properties makes it possible to assess how the unique layered structure of paper affects its performance in use and to speculate on the probable improvements that would result from an appreciable departure from this type of structure. Five main areas of use are considered below.

Printing media

A considerable improvement could be expected from the greater Z-direction strength and resilience. On the other hand, increased permeability to ink could be a disadvantage; the effects on surface smoothness cannot be predicted.

Stiff packaging

The essential property of creasability of boxboard derives entirely from its laminar structure.[39] Boger,[40] however, using the technique of needling, produced non-layered laboratory samples of board with Z-direction strength increased by up to 200 per cent, without any appreciable loss of other strength properties. Unfortunately, that study makes no mention of the effect on creasability. The structure of board is not expected to have an appreciable effect on its stiffness.

Flexible packaging/sack papers

No indications have been found of the probable effects of the layered structure on the ability of paper to absorb energy in stretching.

Hygienic/disposable tissues

The expected small effects of the layered structure on the absorption of liquids and on compressibility are probably unimportant compared with those of, for example, creping. The expected large improvement in tearing and tensile strengths, however, which would result from a non-layered structure at the same degree of bonding, may yield appreciable advantages in the crucial balance of strength and softness.

Barrier/filter media

Again, the small increase in the resistance to the flow of fluids through a layered mat of fibres is likely to be of only marginal direct importance in use. In contrast, the effect of the greater proportion of dead spaces in such a structure on the retention of particles could well be considerable, but difficult to predict until the mechanism of retention is better understood.

Non-layered sheet materials

The unique set of properties of woven cloth are due not just to the three-dimensional interweaving of the threads, but also to their length and regular arrangement, so that a direct comparison with the layered structure of fibres in paper would not be valid. Yet sheet materials exist that consist of fibres that are short and disordered, but oriented in the Z-direction. A striking illustration of such an arrangement of fibres in leather is shown by Kerner.[61] Craik[41] describes how the resilience of human skin results from a similar arrangement and commends this observation to the attention of engineers and materials scientists. Pepper[42] compares natural leather with the 'Corfam' synthetic product and concludes that the decisive advantages of the former stem from its three-dimensional structure.

Attempts have been made to reproduce such structures in artificial products. The ancient industry of woollen felt making uses the anisotropy of the surface friction of woollen fibres, which allows them to mat together into an interwoven, three-dimensional network.[61] The resulting material is tough, flexible and resistant to creasing.

Non-woven materials made from artificial fibres are laid down in a two-dimensional fibre arrangement, but can be given a measure of three-dimensionability by the technique of needling, in which arrays of barbed needles penetrate the thickness of the web and force some fibres to penetrate the different layers. The resulting materials compete successfully with, for example, woollen felts and are also used as bases for artificial leather. Hearle and collaborators[43] present a thorough study of the effects of this technique: it stretches and densifies the fabric and produces an S-shaped stress/strain curve. The strength of unbonded webs may increase up to 7–10 times, depending on the extent of the treatment: but a very high degree of vertical entanglement of the fibres may reduce the strength again, because of disorientation. The modulus and stability of the fabric may be improved also by a prestretching process. Boger[40] also found large increases in the strength of needled laboratory samples of board.

The technique of needling is slow and expensive, with the result that the patent literature lists many descriptions of attempts to simplify the process through using jets of water instead of the needles[44, 45] or jets of gas or steam[46, 47] The variety of other similar attempts demonstrates the interest of the non-woven industry in achieving a three-dimensional structure—by compaction,[48, 49] by forming on a spiked surface,[50] by electrostatic deposition[51] of fibres and even by slitting the formed web, twisting the strips and joining them together.[52] No doubt some of these approaches are not practicable, but others may find application even in the paper industry—for example, compaction.[53]

Attempts to produce a three-dimensional paper structure

THERE are indications that quite straightforward papermaking processes may result in a degree of Z-orientation. It has been mentioned above, for example, that machine-made papers often appear to have higher bulk and greater permeability than corresponding laboratory samples. These effects become more pronounced as the forming consistency increases[54] and are being consciously sought for in the attempts to form at very high consistencies.[9] Claims are also being made that very fast drainage in a direction at an appreciable angle to an inclined wire will cause some fibres to be 'vertically encased in the web'.[55] Formation of a web from foam is also claimed to achieve a measure of Z-direction orientation;[56] this has not been the author's experience.

A completely different approach to the attempts at orienting the fibres in the Z-direction is to include in the furnish a proportion of filler particles, which are themselves of a shape extended in the three directions. These may be multi-axial fibres produced by disintegrating a plastic foam[57–59] or even hollow spheres.[60] Large increases in bulk are claimed, sometimes without a corresponding loss of strength.

Conclusions

A GENERAL survey of the effects of the layered structure of paper and board on its properties in use shows that these effects are either marginal or not detrimental for most conventional uses. A possible exception is the Z-direction strength, which is important for printing applications and which could be expected to benefit considerably from a measure of fibre orientation in that direction.

The requirements are different in the newer applications of paper-like materials, as non-woven replacements of textiles and leather and as fibrous reinforcement of plastics. The layered structure is here a distinct disadvantage; the many and varied attempts to overcome it are discussed.

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Transcription of Discussion

Discussion

Mr B. Radvan May I say first of all that the contributors are to be congratulated, because it is an objective that has been tried before many times, not often with much success. The audience may remember that the Curlator Company, for instance, had a beating machine that claimed to produce curled fibres. More recently, the latency phenomenon in a refiner groundwood was discovered and there again it was found that groundwood fibres that were not allowed to straighten out would produce a sheet that was bulky and soft. There have been many attempts to use crimped fibres. Whether such paper is three-dimensional or not is perhaps an academic question. We are looking forward to hearing of more results.

Miss Sylvia Schmidt We made our tests on a pilot machine in a purely practical way (not as an academic question) and we succeeded in obtaining very well formed webs. Later on, laboratory studies were made at the Reutlingen Textile Institute to find out the best way to use these crimped fibres.

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