Concepts and Evidence of Interdigitation in the Strength Development of Paper and Sheets Formed from Highly Fibrillated Cellulose: A Review

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This review considers published evidence supporting ways in which self-assembly and interdigitated structures that emerge during sheet formation contribute to mechanical strength and structural development in nanopaper and conventional paper-like sheets. Interdigitation is defined here as a form of three-dimensional connectivity within fibrous networks, including elements of parallel interaction, minor weaving, physical entanglement, and inter-diffusion among fibers, fibrils, and nano-scale cellulosic features of multiple length scales. Particular attention is given to out-of-plane fiber orientations and the persistence of three-dimensional connectivity during sheet formation, features that are not fully captured by idealized two-dimensional network models. The review considers mechanical, rheological, microscopic, and processing-related studies to assess how hydrodynamic conditions, flocculation, consolidation history, and drying influence the formation and effectiveness of interdigitated structures. The collected evidence suggests that interdigitation is an inherent feature of papermaking over a broad range of fiber and fibril dimensions, and that it can be strongly influenced by processing conditions. In addition to highlighting the topic of interdigitation, this review also reveals a need for more detailed theoretical consideration, as well as focused experimental work.

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INTRODUCTION

Interdigitation and Self-assembly

The term interdigitation, as used here, refers to a persistent three-dimensional, multiscale arrangement among fibers, fibrils, or nano-scale cellulosic strands. Interdigitation develops during the formation of paper or nanopaper and is preserved during consolidation and drying. Interdigitation can include aspects of overlap, over-and-under features, alignment, and entanglement. It improves adhesion, mechanical coupling, and load transfer within the network, without requiring complete mechanical interlocking. At the molecular and nanoscale levels, this behavior arises from chain mobility, entanglement physics, and interfacial diffusion, which are concepts that are well established in polymer science and captured by reptation theory.

A simple analogy can help visualize this concept: spreading the figures of both hands and bringing them together so that the fingers partially overlap in three dimensions. The fingers are not fully locked together, but the interpenetration restricts movement and increases resistance to separation (Fig. 1). Similarly, in cellulose-based networks, interdigitation strengthens interfibrillar coupling and can enhance the material's overall performance.

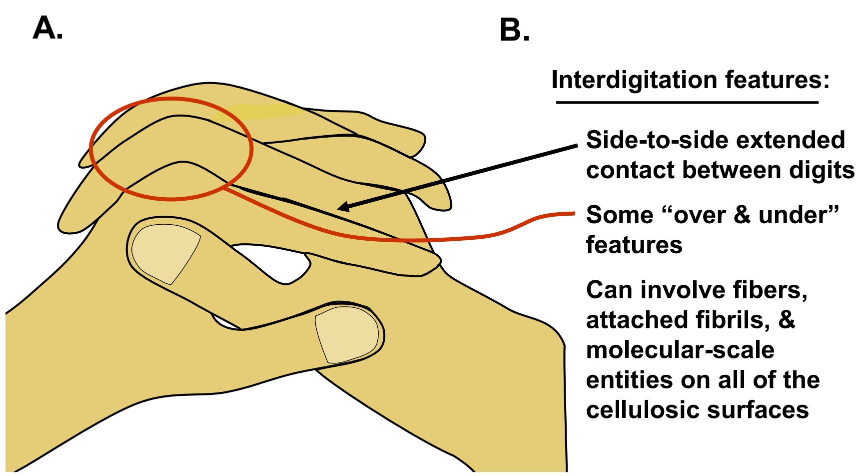

Fig. 1. Sketch of hands joined together with interdigitated fingers (A) along with some defining characteristics of interdigitation, for purposes of discussion in this article (B)

This review considers both the concept of interdigitation and evidence of its importance during the formation of paper and related cellulosic structures. It will be shown that features of interdigitation are present during conventional papermaking, as well as during nanocellulose film formation. It will be argued that aspects of interdigitation among fibers and fibrils are an inherent aspect of ordinary papermaking processes, but the role of interdigitation has not always been appreciated. The connectivity within conventional paper is typically described in terms of statistically distributed fiber crossing and surface contacts within a largely two-dimensional network, arising from fiber flexibility, collapse, and planar deposition during formation (Kallmes and Corte 1960; Deng and Dodson 1994). In contrast, interdigitation emphasizes coordinated three-dimensional cooperation among fibers, fibrils, and nanoscale cellulosic components across multiple length scales. These interactions are made and preserved in the process of sheet formation and drying, and they can result in a mechanically integrated network. Whereas the self-arrangements among fibers within ordinary paper are often described in statistical and geometry-driven terms, interdigitation reflects process-mediated structural organization and exhibits features of hysteresis associated with consolidation and drying. Here the term hysteresis is used to indicate that the forward process of sheet formation and development of bonding within paper (or other cellulose-based structures prepared by dewatering and drying) are somewhat irreversible. At a minimum, higher levels of moisture content will be required to disassemble such structures in comparison to the moisture content at the equivalent point of the forward process.

While such interdigitated features contribute to bonding within paper structures, their broader significance lies in enhanced load transfer, out-of-plane force displacement, and higher delamination resistance (Kallmes and Corte 1960; Deng and Dodson 1994). In general, interdigitation is not simply enhanced entanglement, but an emergent concept of hierarchical self-assembly. This distinction highlights its capacity as a tunable design parameter for sheet architecture and functionality. The present review article mainly considers the fiber scale when seeking examples to demonstrate evidence of interdigitation, with lesser emphasis on the effects of fibrils or nano-scale cellulosic entities. The smaller-scale features are to be considered in more detail in two planned future review articles that will focus to a great extent on some theoretical aspects. A separate future article will focus on product opportunities.

In paper and nanopaper systems, interdigitation naturally occurs because cellulosic fibers and nanofibrils conform to each other and bend around one another during consolidation. These interactions lead to semi-random out-of-plane deviations from the main in-plane orientation, creating under-and-over arrangements that mechanically constrain the network even in the absence of covalent bonds. Although some idealized models assume strictly in-plane fiber orientations (Deng and Dodson 1994; Niskanen 1998), classical theories of paper tensile strength rely on fibers passing under and over each other (Page 1969). When acting across multiple length scales, interdigitation is therefore hypothesized to play an important role in contributing to mechanical strength, viscoelastic behavior, and end-use performance in both conventional paper and nanocellulose films.

Closely related to interdigitation is the broader concept of self-assembly, which refers to processes in which attributes of the system components partially govern how they organize themselves into ordered or functional structures without continuous external intervention. As described extensively by Pelesko (2007), self-assembly requires that each of the components has intrinsic attributes such as shape, surface, or interaction potentials that favor certain arrangements or configuration over others. In cellulosic networks, fiber-like shapes, fiber flexibility, surface functionality, and interfacial interactions collectively promote both self-assembly and the development of interdigitated structures during sheet formation and drying (Mendes *et al.* 2013).

Deviations from an Apparent Dominance of 2D Structure in Paper

As a step towards understanding the role of interdigitation in contributing to the structure and properties of paper or nanopaper, it is hypothesized that various out-of-plane features may be treated as systematic deviations from the commonly assumed two-dimensional model of paper structure. The intent here is not to dispute the main validity of the two-dimensional models that have proven highly successful for explaining many in-plane properties, but rather to use them as a starting point for adding some supplementary features. In this way, out-of-plane interactions such as fiber crossings and partial overlap can be discussed without abandoning the simplicity and explanatory power of established models. The present subsection considers various contributing causes of deviations from ideally two-dimensional structure of paper sheets; later in the article there will be consideration of related effects on paper properties. For instance, there will be a description of the Scott Internal Bond tests and some results of such tests.

Idealized filtration mechanism

In science and engineering, it is common to make predictions based on models that are intentionally simplified. For instance, the ideal gas equation PV = nRT is still very widely used, despite the fact that the molecules or atoms of real gases occupy space and have mutual attractions, which lead to known inaccuracies. Likewise, many in-plane properties of paper, such as tensile strength and elastic modulus, can be adequately understood by imagining an ideal process by which perfectly straight fibers arrive one by one and lie down flat at completely random positions on a forming screen (Deng and Dodson 1994; Niskanen 1998). Within this framework, the tensile forces needed to break paper, as well as the in-plane elastic moduli, can be understood based on a model that does not consider the detailed nature of fiber crossings, including the necessity that one fiber needs to be above or below the other at the point of crossing (Page 1969). As stated by Alava and Niskanen (2006), "a two-dimensional random fiber network approximation is good for many purposes." Nevertheless, such models necessarily omit features that may become important when considering thickness-related phenomena, stress transfer at crossings, or the development of interlocking and constraint in the out-of-plane direction.

As a first step in building on such a model, it will be assumed that a set of fibers, which are initially straight, non-contacting, and at various orientations within a suspension are being formed into paper under simple laminar flow conditions. In an attempt to draw attention to the hypothetical cellulosic fibers, they are drawn as colored cylinders. By contrast, the forming fabric image represents a section of a commercial forming fabric. In Part A of Fig. 2, three such fibers are envisioned as approaching the forming fabric of a paper machine. Once each fiber first impinges upon the forming fabric, it is expected to rotate so as to lie down in an approximately horizontal position.

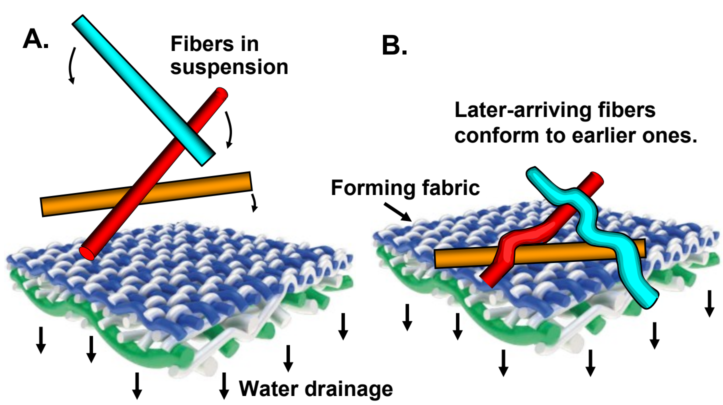

Fig. 2. Conceptual illustration of (A) variously oriented fibers in a suspension being drawn toward a paper machine forming fabric by downward flow, followed by sequentially landing on the forming fabric, rotating approximately into the plane of the sheet, and bending to accommodate any earlier-arriving fibers

As shown in Part B, later-arriving fibers are shown as bending around any preceding fibers to accommodate their presence. Such bending can be reasonably expected, especially in the case of refined kraft fibers, since internal delamination within the cell walls is known to render them highly conformable in the wet state (Baker 1995; Batchelor *et al.* 1999). Although the fibers shown in Fig. 2 cannot be properly described as being "woven" in the conventional sense, even a process of sequential arrival has the potential to create some rudimentary over-and-under configurations. These features represent an early form of out-of-plane engagement that may be viewed as a precursor to interdigitation, providing a physical basis for introducing three-dimensional constraints into otherwise two-dimensional network models.

Out-of-plane orientation of fibers

The illustration just shown requires that at least some of the fibers arriving at the forming fabric will have markedly out-of-plane orientations. This follows from the widespread practice of maintaining a small difference in velocity between the jet of fiber suspension and the forming fabric or pair of fabrics (Niskanen and Hämäläinen 2012; Vahey *et al.* 2013). When the jet impinges onto the first fabric surface, a shear field will develop, causing any individual fibers to undergo end-over-end rotations (Mason 1954; Phan-Thien 2016). It is well known that fibrous particles within suspensions tend to align themselves in response to shear flow (Hubbe *et al.* 2017). Analogous effects are induced during some traditional handmade papermaking practices, wherein the hand papermaker uses a "shake" or delicate "tilting" methods while allowing water to drain through a screen (Hunter 1947; Hubbe and Bowden 2009). Although these processes differ in scale and flow complexity, both introduce transient shear fields that can rotate fibers out of the plane prior to deposition. Taking the model of simple, laminar shear as a simplified model of the situation, the rotation speed is expected to be fastest when the fiber is strongly out-of-plane relative to the imposed shear, but it will be low at moments when the fiber is close to being aligned with shear (Phan-Thien 2016). This effect is illustrated in Fig. 3.

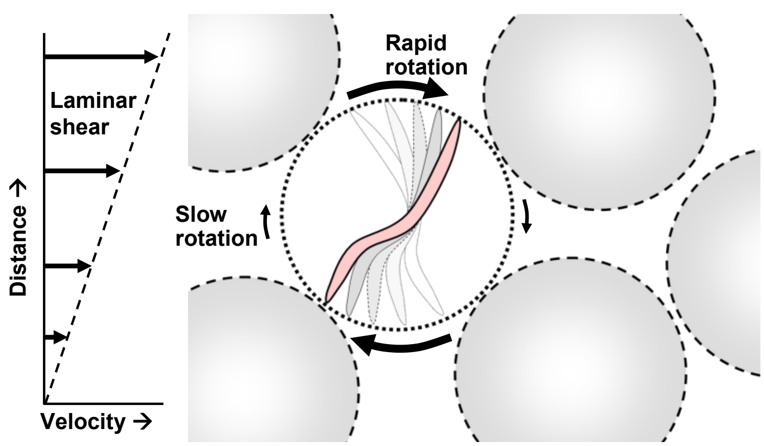

Fig. 3. Concept of fiber rotation in laminar shear flow, which gives rise to preferential machine-directional fiber orientation due to jet-to-wire speed differences during paper formation, but which also implies that some fibers will have an out-of-plane orientation within the paper

The uneven rotational speed of individual fibers under shear, as illustrated in Fig. 3, is part of the explanation for papermaking fibers to show a preference of machine-directional orientation in typical sheets of machine-made paper (Alava and Niskanen 2006; Vakil *et al.* 2010; Niskanen and Hämäläinen 2012). Observations of this type have been predicted by simulations of fiber suspension flow (Lindström and Uesaka 2008). Such alignment also has been shown to correlate with higher strength in the machine direction of most machine-made paper sheets (Gigac and Fiserová 2010). What is often overlooked in such discussions is the fact that the same rotational motions giving rise to preferential machine-directional fiber orientation also entail an assumption of out-of-plane orientation of at least a portion of fibers during processing. Direct experimental evidence of such "fiber tilt" was provided by Vahey *et al.* (2013) in a study aimed at accounting for why "tape peel" tests often give different results depending on the "forward" or "backward" machine-directional orientation of the tests.

A more extreme depiction of the same kind of situation was shown by Niskanen (1998), in which one of the fibers was shown to span about three layers of predominantly two-dimensional layering in a hypothetical sheet of paper, emphasizing the potential for localized three-dimensional connectivity within an otherwise planar network. Figure 4, which is redrawn based on the concepts behind original drawings in Niskanen's (1998) monograph, contrasts the cross-section of a strictly two-dimensional paper sheet (Part A) with a sheet in which some of the fibers span two or three different layers within the structure (Part B). The term "felted" is sometimes used when referring to such an internal structure within a paper sheet. By contrast, the sheet represented in Fig. 4(A) can be called "layered". In addition, the same publication depicts the most rudimentary form of entanglement (Part C).

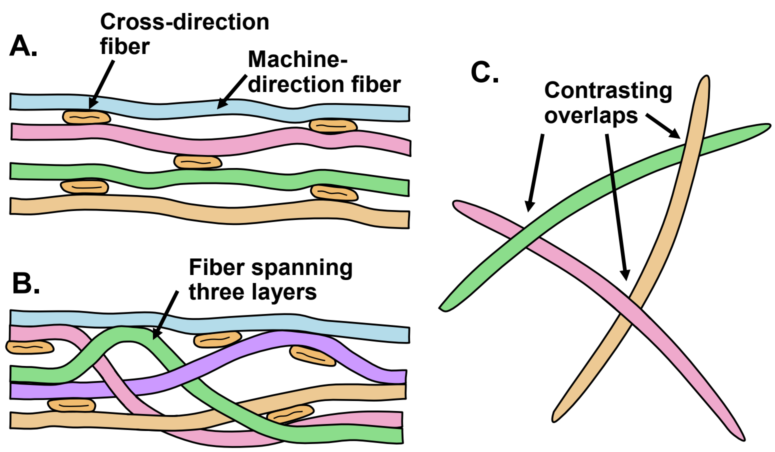

Fig. 4. Redrawn illustration from Niskanen (1998) depicting: (A) the cross-section of a hypothetical paper sheet that was formed in a strictly two-dimensional layered manner; (B) a sheet in which at least fibers bend to span either two or three layers of the hypothetical paper sheet, and (C) Niskanen's example to define the most rudimentary form of entanglement

Due to the high aspect ratio (*e.g.* about 100) of typical papermaking fibers, it can be challenging to obtain an actual cross-sectional image of paper that could provide support for the hypothetical structure shown in Fig. 4(B). Figure 5 shows such an image, which comes from the work of Dickson (2000). The label in the upper right of the figure has two red arrows pointing to the cross-sections of two fibers that are clearly facing approximately perpendicular to the plane of view. Note, for instance, the presence of a lumen space in the center of each of the indicated fibers. The label at the lower right of the figure has an arrow pointing to a fiber that was at least partly somewhat aligned with the plane of view. Notably, the latter fiber spans from approximately the second layer of fibers (starting from the left) to the third or fourth layers of fibers at the right of the figure.

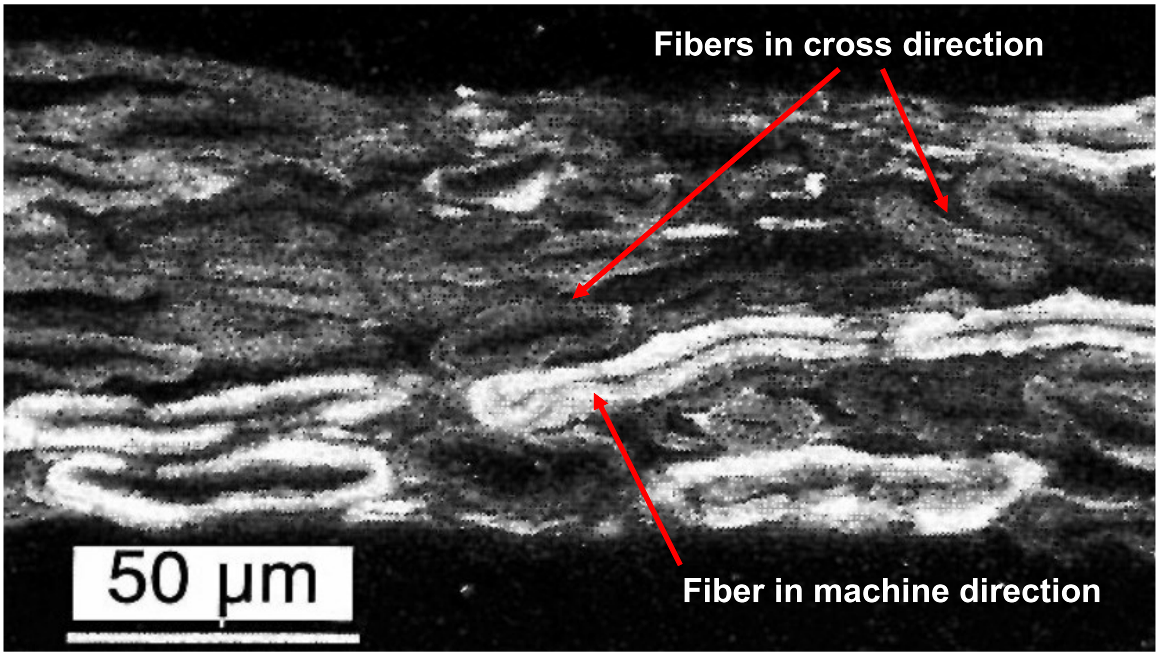

Fig. 5. Scanning electron micrograph of a paper cross-section, as reported by Dickson (2000). The two labels with arrows have been added. Permission to use the image was obtained from APPITA, the publisher.

Another source that can be used to seek evidence for out-of-plane orientation of fibers within paper structures is X-ray microtomography (Viguié *et al.* 2013). The cited authors obtained images within which different fibers were individually shown with contrasting colors. Evidence of fibers spanning more than one plane within the structure were clearly apparent in sheets that had been formed from copper fibers. However, in the case of cellulosic fibers formed into paper handsheets, the fibers were mainly restricted to horizontal plains. Such results are consistent with the highly dilute fiber suspensions that are employed in standardized laboratory forming procedures used for paper testing. Such procedures inherently favor one-at-a-time arrival of fibers onto the top of a fiber mat, as was illustrated schematically in Fig. 2.

It should be noted that the conceptual model assumed in the drawing of Fig. 4 did not take account of the fact that the flow environment can be expected to impact the shapes of flexible fibers in the suspension. In reality, hydrodynamic forces can bend and curve fibers during transport and deposition, further complicating their final spatial configuration (Mason 1954). Figure 6 depicts some representative shapes of fibers that were captured by high-speed photography in the cited work, illustrating the diversity of conformations that can arise under realistic flow conditions.

Another way to account for a somewhat three-dimensional structure of fibers within a paper sheet has been described using the word "thickening" (Norman 1989). In contrast to the filtration mechanism where fibers are assumed to deposit individually onto a forming surface, the thickening mechanism envisions the fibers maintaining a sort of network structure during removal of water (Steenberg *et al.* 1965). As dewatering proceeds, the pre-existing network is progressively compacted rather than being fully dismantled and reassembled.

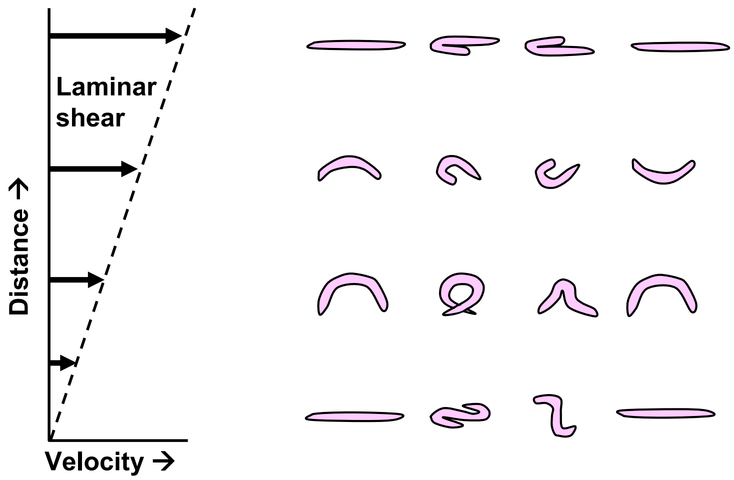

Fig. 6. Representative shapes of pulp fibers in laminar shear flow, as observed by Mason (1954). Redrawn

Such a mechanism might be argued to be consistent with the multiple pulses of vacuum and spring-back cycles as a paper sheet passes over multiple hydrofoils, vacuum boxes, and press nips on its way towards entering the evaporative drying stage (Hubbe *et al.* 2020). These cyclic loading and unloading events provide repeated opportunities for a fiber network to persist, deform, and recover, rather than collapsing into a purely planar arrangement at each stage of water removal. The thickening model is also consistent with the fact that most commercially made paper is formed from fiber suspensions that have a high enough crowding factor to favor frequent and sustained interactions between fibers (Kerekes and Schell 1992). Figure 7 illustrates how a thickening-type process can be expected to result in cases where the crowding factor within a suspension is high enough to result in a shear-tolerant, space-spanning fiber network involving essentially all of the fibers that are present.

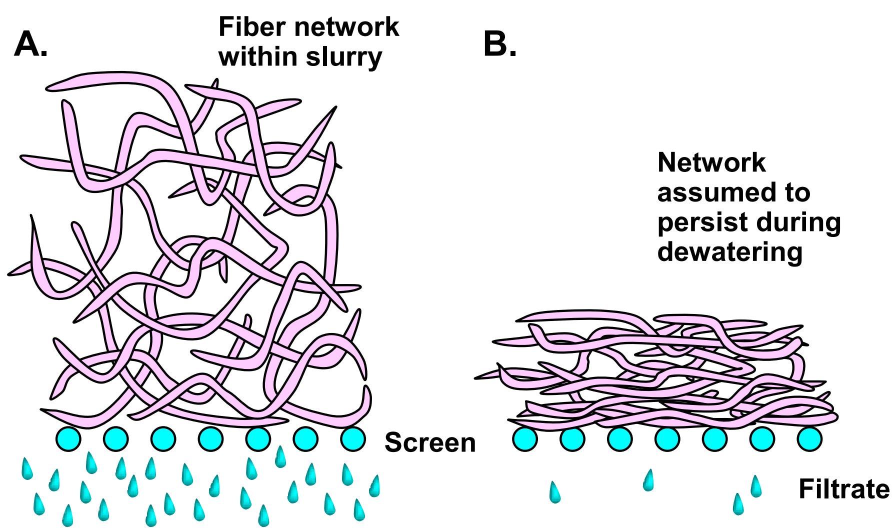

Fig. 7. Conceptual illustration of a paper sheet being formed from a fiber suspension in which a network is already present due to a sufficiently high solids content, as well as high enough aspect ratios of the fibers

Presence of fiber flocs in agitated suspensions

Consistent with the preceding discussion of out-of-plane fiber orientation, and the persistence of three-dimensional connectivity during formation, some of the earliest manifestations of such behavior can be seen in the formation of fiber flocs within papermaking suspensions. Examples of hypothetical primitive fiber flocs, which readily form in stirred suspensions of papermaking stock, are shown in Fig. 8 (Parker 1972).

Fig. 8. Depiction of some simple fiber flocs in which just a few fibers may be held together by a combination of fiber-to-fiber friction and elastic forces of straightening after fibers have become momentarily bent by eddies of flow (examples redrawn based on originals by Parker (1972)

Because such flocs can be held together by the elastic forces within the fibers themselves, along with friction, it is clear that such flocs require at least a minor amount of interweaving. It has been noted that more intense or more extensive floc formation can be induced by addition of various flocculating chemicals to agitated fiber suspensions, thereby further reinforcing fiber-fiber connectivity (Hubbe 2007; Kozel *et al.* 2025). There is evidence that fiber flocs present within the suspension before formation of the paper web can maintain or retain at least part of their structural integrity as the paper sheet is being formed. Such a hypothetical process is illustrated, in simplified fashion, in Fig. 9. Although it can be expected that the structures of real fiber flocs in a papermaking suspension will tend to be more complicated, one of the simple examples from Fig. 8 is being used here to illustrate a possible mechanism. Note that the floc example shown in part A is the same as the middle example shown in the previous figure and that two copies of the same floc are pictured as becoming a part of a hypothetical paper sheet in part B of the figure.

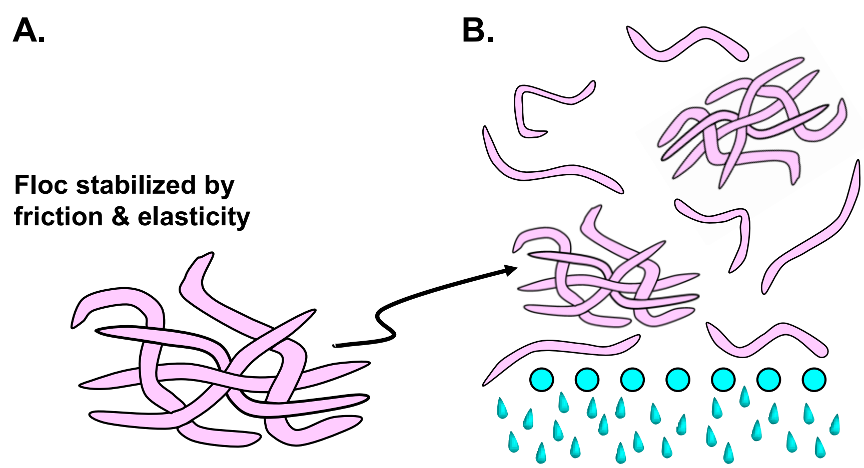

Fig. 9. Proposed persistence of fiber floc structures present in an agitated suspension (part A) during the formation of a paper sheet (part B) on a screen (shown as cyan-colored cross-sections of strands perpendicular to the plane of view).

Nanocellulose and interdigitation

Though, as already mentioned, the present article mostly considers examples having to do with ordinary papermaking, it can be anticipated that structures formed from some nanocellulose products, such as NFC, could have relatively high levels of interdigitation. Contributing factors suggesting the possibility of high levels of interdigitation include a relatively high ratio of length to width in typical NFC material and relatively high flexibility of narrow fibrils. In addition, some interesting observations, possibly related to interdigitation, came to light in some recent research related to the usage of NFC as an additive during the preparation of paper. Specifically, there is a need to better understand some recent findings related to the use of nanocellulose as a dry-strength agent during the production of paper (Barrios *et al.* 2023; Hamm *et al.* 2024; Jones *et al.* 2024; Atree *et al.* 2025; Kozel *et al.* 2025). Findings from that series of studies suggest that nanocellulose can, under certain conditions, exhibit an "antisocial" character, meaning that it resists forming effective bonding with adjacent cellulosic fibers during the drying of a paper sheet (Hubbe 2025). A more technical term such as "not available for interdigitation" might be used.

Such findings prompt the question of whether or not there has been sufficient consideration of interdigitation as an important contribution to the strength of either ordinary paper or paper made with the addition of NFC to the fiber suspension. A key challenge in answering such questions is that it is quite difficult to observe or quantify such phenomena. As a consequence, one is often left with having to rely on evidence or correlations with sheet properties or other secondary evidence, such as mechanical performance, rheological behavior, or other structural proxies, rather than being able to directly quantify the interdigitation itself. Despite this limitation, the authors hope that, maybe as a consequence of the present initial attempt to review this area of technology, there will be additional research progress that can provide a fuller understanding in coming years. Accordingly, the present review considers a range of experimental observations and indirect evidence suggestive of interdigitation phenomena, with particular emphasis on systems involving cellulosic fibers or nanocellulose materials and on how such three-dimensional interactions may contribute to strength development in paper and related fibrous networks.

Concept of an Interdigitation Index

Though much of the evidence to be considered in the present article is qualitative, there may be value, in the future, to attempt to express the extent of interdigitation in quantitative terms. It is proposed here that an interdigitation index (I_i) might be expressed in the following general form:

$$I_i = [\Sigma\,(f_{side\text{-}to\text{-}side}) + \Sigma\,(f_{under\ \&\ over}) + \Sigma\,(f_{spanning\ layers}) + \Sigma\,(f_{entanglement}) + etc.]\ /\ (\text{number of qualified units}) \quad (1)$$

In this expression, the term ($f_{side\text{-}to\text{-}side}$) refers to countable instances of side-to-side features (defined in rudimentary form in Fig. 1), ($f_{under\ \&\ over}$) refers to instances of under-and-over features, ($f_{spanning\ layers}$) refers to instances in which a given fiber occupies more than one definable layer in a paper structure, and ($f_{entanglement}$) refers to a unit of entanglement in the paper-like structure. All of the summation terms in the numerator, added together, are divided by the number of structural units (*e.g.* papermaking fibers) that are being considered in the calculation. For instance, this form of Eq. 1 mainly considers features that could be important for fiber-scale calculations, which means that a fiber could be designated as the qualified unit for such calculations.

The possible future usage of Eq. 1 can be illustrated by considering the "folded hands" iconic example from Fig. 1. In that case, depending on exactly how the terms are defined in future work, one might calculate as follows: In the numerator, suppose that there are nine side-to-side features (meaning that the adjacent fibers are lined up so that they are in contact over a sufficient length to meet the selected qualifications) and four under-and-over features. The "number of qualified units" would be two, since there are two folded hands. Thus, the calculation could be as follows: $I_i = (9 + 4)/2 = 6.5$.

EVIDENCE OF SELF-ASSEMBLY OF CELLULOSE FIBERS OR FIBRILS

Based on a search in the literature, evidence of self-assembly of cellulosic fibers, fibrils, or smaller hair-like groups of cellulose polymer tails can be grouped into different but complementary categories. One of the biggest sources of evidence arises from the measured properties of paper itself and the ways in which those properties depend on fiber characteristics, processing conditions, and sheet structure. Following a discussion of such property-based evidence, subsequent topic areas will include evidence related to certain papermaking processes, the formation and behavior of nanocellulose-based structures, rheological properties of cellulose-containing suspensions, and clues related to the biosynthesis or regeneration of cellulose from solution. Taken together, these diverse sources provide converging support for the occurrence of spontaneous or partially directed organization phenomena in cellulosic systems. As will be shown, much of the evidence tends to be indirect and qualitative in nature. But on the other hand, there are multiple sources of evidence from past studies, and such evidence can be used to support self-consistent concepts related to interdigitation.

Evidence from the Properties of Conventional Paper

In-plane properties of paper

The properties of typical sheets of paper can offer a useful, albeit indirect, window into their internal organization. Several lines of evidence suggest that paper cannot be fully described as a simple two-dimensional network of fibers. Some of this evidence emerges from the degree to which paper's mechanical properties can or cannot be simulated by use of two-dimensional structural models. Further evidence is provided by measurements of strength in the thickness (z) direction, *i.e.*, testing involving delamination or internal bond tests, which probe interactions between fibers across layers. Microscopic examination of fracture surfaces following delamination or tensile failure can further reveal aspects of fiber interlocking or pull-out that are consistent with interdigitation. Finally, there have been some studies attempting to find a relationship between the external fibrillation of fibers during mechanical refining, and the resulting inter-bonding of the fibers and paper strength, thereby providing clues about the role of fibrillar-scale intertwining or interpenetration.

In the future, as researchers focus more deeply into the mechanistic details and practical consequences of interdigitation, it might be possible to consider a wide range of paper strength characteristics as sources of evidence, even when considering some tests that at first seem to involve just two dimensions. For example, it would make sense that the crush resistance of paperboard might depend on resistance to delamination and thereby have some relationship to the focus of this review. This is because failure of a sheet during a crush test might be initiated by local delamination. By contrast, it is reasonable to expect the tensile strength and modulus characteristics of paper to show fewer attributes that can be definitively attributed to features that go beyond a two-dimensional structural model of paper.

Fibrous or cylindrical materials possessing sufficiently high aspect ratios are able to engage in three-dimensional interdigitated packing structures during network formation. In general, systems with relatively low aspect ratios ($L/D \leq 10$) behave analogously to anisotropic granular particles, where contacts are primarily local and dominated by steric constraints. As the length-to-diameter ratio increases into intermediate regimes ($L/D \sim 10$ to 10^2), the probability of multiple simultaneous contacts per fiber rises in a non-linear manner, which causes a transition from particle-like packing to percolated networks, as predicted by excluded-volume and continuum percolation theories for elongated objects (Balberg *et al.* 1984; Kyrylyuk and van der Schoot 2008; Sahimi 1994). At sufficiently high aspect ratios ($L/D \geq 10^2$), increasing fiber length relative to a characteristic pore dimension of the growing network enables penetration and bridging across multiple domains of fibers, thereby promoting out-of-plane contacts and hierarchical mechanical interlocking.

Figure 10 emphasizes some characteristic differences in the manner in which clusters of cellulose entities are likely to be held together as persistent structures, depending on the range of aspect ratios. As mentioned before, rudimentary flocs present in papermaking systems, especially those observed in the absence of retention aids, are expected to be held together by a combination of elastic restorative forces (tending to straighten the fibers) and friction at points of contact. By contrast, such features as twisting (*e.g.*, during the formation of strands) and loop-type entanglements would become common only at much higher values of aspect ratio.

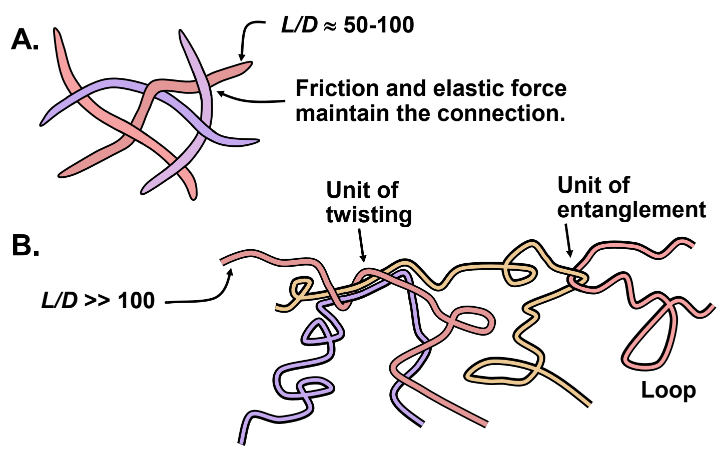

Fig. 10. Conceptual illustration of different ways in which clusters of cellulose-based entities may be held together in aqueous suspensions, depending on the ranges of aspect ratio: A. Expected governing factors for papermaking fibers; B. Some expected governing mechanisms for fibers with much higher aspect ratios, *e.g.* TEMPO-oxidized NFC

The interdigitating network reflects a probabilistic shift in network topology. The influence of particle anisotropy on suspension structure and connectivity is fundamentally entropic in origin. Excluded-volume theory indicates the number of intersections among elongated objects linearly scale with aspect ratio, in essence reducing the percolation threshold and increasing the average coordination number (Onsager 1949; Balberg *et al*. 1984; Sahimi 1994). Thus, fibers with sufficiently high *L*/*D* (≥ 10^2) do not "rest" against one another; rather these materials weave, overlap, and interdigitate in a manner that leads to three-dimensional connectivity within the network. This activity appears to be favored by entropic considerations; by gaining contacts, each fiber, as in the case of a "V"-shaped flock of birds, achieves a minimum in its relative instability and exposure. Such activity does not require chemical bonding or adhesion. Instead, this formative state emerges from geometric constraints, hydrodynamic distributions during formation, and consolidation, which tend to reduce free volume and promote fiber penetration into neighboring void spaces (Deng and Dodson 1994).

While interdigitation may coexist with fiber entanglement and inter-fiber bonding, it is distinguished here as a geometrical and spatial interpenetration phenomenon. Entanglement describes random crossings, but interdigitation explicitly describes 3D penetration and structural integration that survives densification (Onsager 1949; Deng and Dodson 1994). In fibrous sheet consolidation, fluid flow, flocculation, and consolidation history influence orientation and pore evolution (Deng and Dodson 1994). When *L* > evolving pore scale during drainage and pressing, the probability of through-thickness bridging goes up, producing a mechanically percolated network irreducible to simple two-dimensional network models structures (Balberg *et al.* 1984; Kyrylyuk and van der Schoot 2008; Sahimi 1994). The 3D connectivity can be expected to contribute to stiffness, strength, and resistance to delamination, especially as drying freezes these interpenetrating structures (Deng and Dodson 1994).

Thus, fibrous materials above a critical aspect ratio adopt interdigitated 3D packing structures. This onset depends not just on *L*/*D* but on flexibility, friction, and kinetics of network consolidation. From this perspective, strength development in fibrous assemblies is not merely a consequence of increased contact density, but of a topological transition toward hierarchical, 3D interdigitation propelled by aspect-ratio-weighted topological constraints.

One of the most serious challenges that needs to be recognized in any discussion of interdigitation is the fact that the best-known theoretical descriptions of paper's strength have been based on two-dimensional representations of fiber networks.

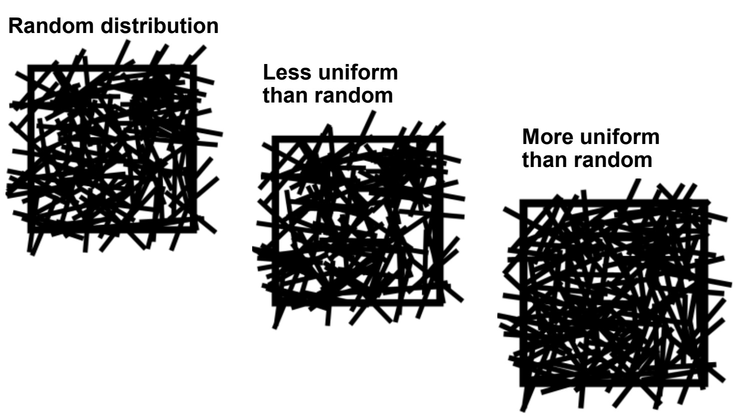

Fig. 11. Illustrative examples of two-dimensional models of paper structure, comparing a purely random distribution (with locations determined by random number generator), *vs.* examples in which the formation was either worsened or improved by moving each fiber a set number of pixels closer to the nearest neighbor or the same distance toward being evenly spaced

As an illustration of the general 2D approach, the locations and orientations of fibers within paper have sometimes been represented by completely random arrangements (Deng and Dodson 1994). This kind of approach is illustrated in Fig. 11, for which a random number generator was used by the present authors to determine the placement of fibers, which were assumed to be completely straight and all the same length. The "less uniform than average" example was created by moving each fiber closer to its nearest neighbors by a set number of pixels. Likewise, the "more unform than average" example involved moving each fiber by the same number of pixels to be more evenly spaced with its nearest neighbors.

The notable success of such models as the Page equation (Page 1969) in being able to explain some of the most important attributes of paper, especially strength properties measured within the plane of the sheet, might initially suggest that out-of-plane interdigitation of fibers within paper plays only a secondary role in paper strength development. This apparent adequacy of two-dimensional models therefore represents both a strength and a limitation: while they capture dominant in-plane mechanisms, they may obscure subtler three-dimensional contributions. At the same time, several well-established observations point to structural organization that arises naturally during papermaking and that is consistent with aspects of self-assembly or directed assembly.

As a starting point for proposing an important role of interdigitation, it has been widely noted that machine-made paper sheets often have a much higher breaking strength in the machine direction as opposed to the cross direction (Hansson *et al.* 1989; Niskanen 1998; Vakil *et al.* 2010; Niskanen and Hämäläinen 2012). Strong correlations have been shown between the predominant orientation of fibers and the relative strength of paper in those directions (Gigac and Fiserová 2010; Kouko *et al.* 2014; Rech *et al.* 2021). For instance, Htun and Fellers (1982) achieved MD/CD ratios of tensile stiffness in the range of about 1.4 to 5.3 depending on the degree of MD-preferred orientation imparted during dynamic formation of paper in the laboratory, followed by unrestrained drying. Those results are shown replotted in Fig. 12.

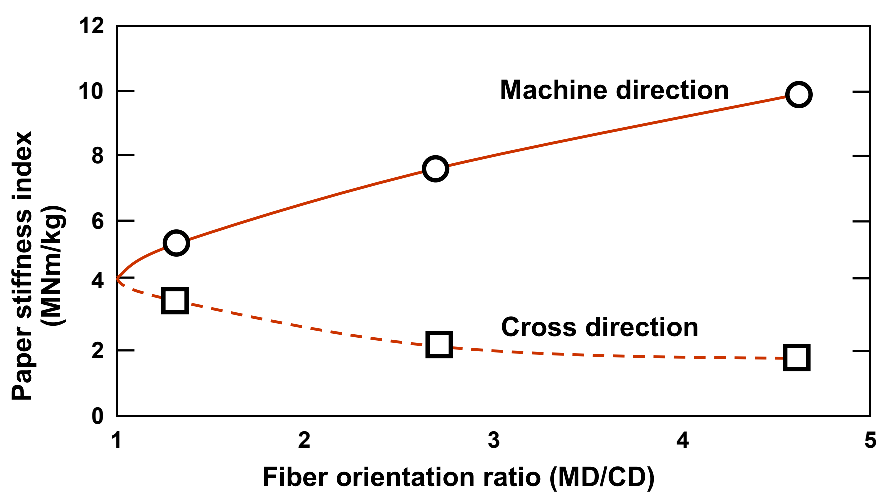

Fig. 12. Dependency of paper's in-plane tensile stiffness relative to average in-plane orientation of the fibers (replotted based on original by Htun and Fellers 1966)

In a way, evidence such as that shown in Fig. 12 provides direct evidence of at least one contribution to interdigitation that results from a key unit operation of papermaking – the tendency of papermaking fibers to become aligned in machine direction both as they exit the headbox slice, and also as they respond to jet-to-forming fabric velocity differences promoting preferential alignment of fibers in the machine direction (Gigac and Fiserová 2010; Vahey and Considine 2010; Vahey *et al.* 2013). Such alignment can be attributed to processes that are only indirectly controlled by those in charge of papermaking operations. Such alignment allows fibers in the paper to have more side-to-side contacts with their neighbors over longer distances, thus strengthening the paper in the direction of production. An additional kind of anisotropy is a strong tendency for most fibers within paper to be mainly oriented within the plane of the sheet, reflecting both gravitational settling and hydrodynamic flattening during consolidation (Radvan *et al.* 1965; Radvan 1973; Niskanen 1998). Such behavior is contrary to what the fibers in a stochastically random environment favor, *i.e.* adoption of orientations in which they form a percolation network to maximize contact points.

The points just mentioned give rise to an important question: At what point can the alignment of fibers be regarded as qualifying as an aspect of interdigitation? Here it will be proposed that the line of demarcation can be drawn based on parallel alignment between pairs of adjacent fibrillar units for sufficient length, as selected by future researchers. For instance, there might be a rule that two adjacent fibers or fibrils need to be in side-to-side contact over a distance that is at least ten times their characteristic diameter. Thus, rather than focusing on the average orientation, attention is directed toward neighbor-neighbor arrangements.

Despite the widespread acceptance of two-dimensional models to account for paper's strength properties, there has been at least some realization that such models are insufficient to provide a complete accounting of paper's observed properties. Kappel *et al.* (2009) stated that models based on a strictly two-dimensional structure of paper do not correctly account for the development of relative bonded area within paper. For instance, such models ordinarily do not consider development of side-to-side bonding interactions between the vertically oriented sides of fibers in a paper sheet. Further supporting this view, Vahey and Considine (2010) showed that jet-to-fabric velocity differences, in addition to tending to align fibers in the machine direction, also give rise to out-of-plane orientation, thereby helping to resist delamination of the paper, as discussed below. These findings imply that three-dimensional fiber arrangements arise naturally during papermaking and may play a more significant role in thickness-direction properties than is typically acknowledged.

Resistance to delamination

Out-of-plane attributes of paper strength are of key importance when assessing whether or not interdigitation phenomena play an important role in the development of paper properties. Particularly, resistance to delamination provides an indirect but sensitive probe of interactions that extend across layers of fibers. Conceptually, at least three mechanisms can be envisioned by which out-of-plane arrangement of fibers may arise during papermaking and contribute to delamination resistance. The first is based on a simple filtration-based mechanism, whereby fibers mainly arrive one-by-one at the forming screen (or at the top of a mat of previously deposited fibers). As had been illustrated in Fig. 2, as water continues to drain towards the screen, incoming fibers tend to lie down in a more-or-less horizontal manner, coming to rest either directly on the screen or on an earlier-arriving layer of fibers. However, when such a fiber attempts to lie down, it will tend to bend in order to conform to the shapes of fibers already present in the preceding layer of fibers. This geometric constraint can force fibers to bend or partially wrap around underlying fibers, thereby creating local out-of-plane contacts that may enhance mechanical interlocking between layers. Second, eddies of flow within the suspension, such as hydrodynamic disturbances resulting from jet-to-fabric speed difference, can be expected to induce an alignment of some of the fibers in an out-of-plane organization prior to deposition. This had been illustrated in Fig. 6. Fibers subjected to such flow conditions may arrive at the forming surface with significant angular deviations from the sheet plane. If a transient fiber network structure exists in the suspension, these fibers may be prevented from fully relaxing into a two-dimensional orientation upon their arrival at the top of the fiber mat, thereby preserving some degree of out-of-plane alignment within the consolidated sheet. Third, many of the fibers may be incorporated into persistent flocs within the suspension (Kerekes and Schell 1992; see Fig. 8). Such flocs can contain fibers that are already mechanically constrained in three-dimensional arrangements prior to sheet formation. When these structures are immobilized during dewatering, they may introduce felt-like regions within the sheet that contribute to resistance against layer separation. A combination of such contributions is expected to give rise to the kind of layer-crossing fibers as were illustrated in Fig. 4B and documented by Vahey and Constidine (2010).

It is well known that paper's Z-directional strength is often low in comparison to its in-plane strength, when normalized to account for sample dimensions (Radvan 1973). However, it has been reported that single-ply paperboard products, which have higher mass per unit area, tend to have a greater degree of out-of-plane fiber orientation (Niskanen *et al.* 1997). This effect has often been attributed to the common use of higher consistencies of fiber slurries during forming, which favors the development of persistent flocs. Increased flocculation would be expected to promote more three-dimensional fiber arrangements, thereby giving a more felt-like internal structure to the resulting product. Although these concepts help to support the expectation of enhanced out-of-plane structures, especially in paperboard grades, direct evidence linking such structures to measurably improved delamination resistance remains limited. This gap highlights the difficulty of isolating interdigitation effects from other contributions to Z-directional strength, such as increased bonding area or densification.

Evidence of effects due to out-of-plane orientations of fibers can be sought in the results of delamination strength tests and post-failure observations. Some studies have reported that delamination or internal bond failure results in dangling ends of fibers or fibrillar features extending outwards from the plane of delamination (Robinson 1980; Stratton and Colson 1993; Kang *et al.* 2004; Schmied *et al.* 2013; Hirn and Stennach 2025). Such features suggest that some fibers or fibrils had spanned the delamination interface prior to failure, consistent with partial interdigitation.

Figure 13 illustrates how such raised fibers often can be readily observed after the completion of the Scott internal bond test (TAPPI Test T541). As shown, the test is carried out using a weighted pendulum, which loses some of its energy when it strikes an aluminum piece having a right-angle bend. The impact causes delamination of a paper specimen that is held in place by means of strong double-sided tape on both sides. A higher level of Z-directional bonding results in absorption of more energy, thereby decreasing the maximum point in the pendulum's swing, which is recorded by the device.

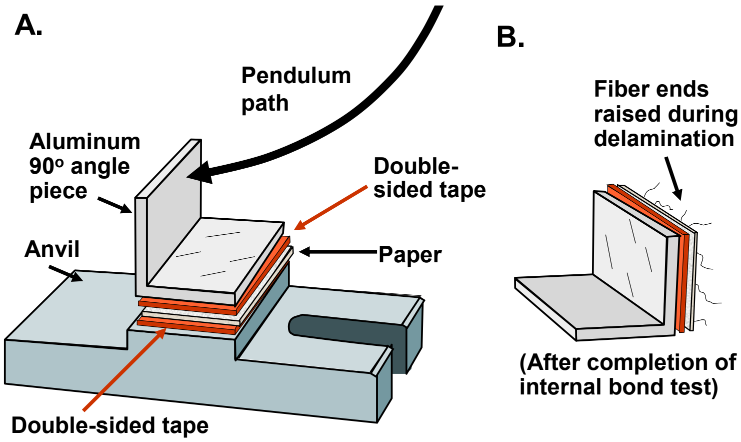

Fig. 13. Illustration of TAPPI T541 test procedure, which can give rise to raised fibers in the course of delamination of paper sheets: A. Apparatus and set-up; B. Commonly observed state of raised fibers at the completion of test

The work of Stratton and Colson (1993) is particularly noteworthy due to its controlled experimental design. In their study, pairs of refined softwood kraft fibers were allowed to dry in contact and subsequently subjected to carefully controlled shear-detachment prior to preparation of scanning electron micrographs. Most of the extended hair-like fibrils visible on the cellulose fiber surfaces after bond breakage were about 10 μm or less in length and less than 1 μm in diameter. While such observations do not conclusively demonstrate pre-existing interdigitation, they provide suggestive evidence that fibrillar scale features may bridge fiber interfaces and contribute to resistance against separation.

Microscopic evidence in general

There has been little reported work attempting to directly document evidence of interdigitation prior to the breakage of fiber bonds in paper, reflecting the experimental challenges associated with visualizing such features in intact paper. Nanko and Ohsawa (1989) employed a differentially staining approach, using two sets of refined hardwood bleached kraft pulp with different types of colloidal metal spheres. It had been established that such spheres stick to external surfaces of fibers and fibrils without permeating into fiber cell walls. By varying the extent of refining, wet-pressing conditions, and drying protocols, including whether the web was air-dried or dried after solvent replacement to avoid dimensional changes, *e.g.* shrinkage, the authors sought to preserve the native structure of fiber-fiber contacts. Following this, the dried structures were impregnated with epoxy resin, which allowed subsequent preparation of ultra-thin cross-sectional specimens. The main finding of the work was that secondary cellulosic fines (created during refining) formed a thick layer adjacent to the bonded areas between fibers, thereby appearing to contribute to bonding. However, the study did not reveal any evidence of fibrils attached to opposing fibers becoming intertwined with each other. Instead, such fibrils were present in a very thin layer between adjacent fibers in the specimens from the impregnated paper. It is important to note that the investigation focused primarily on the intermediate size range of fibrils, as well as cellulosic fines, leaving open the possibility that finer, molecular-scale features could contribute to interdigitation but remain below the resolution of the imaging technique.

More direct quantification of evidence of fiber orientation within paper was obtained by Enomae *et al.* (2008), who used confocal laser scanning microscopy. The fibers had been stained with fluorescent dyes. They found that wood-containing fibers, which tend to be stiff, with a high frequency of kinks and curls, showed high frequencies of out-of-plane fiber orientations. In contrast, copy paper, which is composed largely of well-refined fibers, showed a predominantly in-plane orientation of the fibers. Schmidt (1973) showed that intentional crimping of fibers led to paper sheets with markedly enhanced three-dimensional fiber orientation. These observations support the view that processing-induced fiber morphology can strongly influence the degree of out-of-plane organization, thereby affecting the potential for interdigitation and resistance to delamination.

Fibrillation vs. paper strength

There has long been disagreement among investigators as to whether or not external fibrillation at the surfaces of refined kraft fibers makes a significant contribution to the development of paper strength. Much of this disagreement can be traced to differences in how fibrillation is defined, how it is generated, and which strength properties are emphasized in different studies. Clark (1942, 1978, 1984) assigned a high importance to external fibrillation, arguing that fibrils extending from cellulosic fiber surfaces provide a highly effective surface area and contribute high flexibility in the wet state, thereby facilitating development of extensive bonded area between adjacent fibers as the sheet dries and contributing directly to strength. This interpretation implicitly assumes that surface-attached fibrils participate actively in fiber-fiber contacts and that their presence translates into mechanically effective bonding. Emerton (1980) took an opposing view, arguing that strong inter-fiber bonding can develop between cellulosic fibers even in the absence of apparent fibrillation, emphasizing instead the importance of fiber conformability and internal structural changes within the fiber wall. This latter view is supported, at least in part, by observations that the resulting strength of paper is often not greatly affected by treatment of refined pulp with cellulolytic enzymes (Bhardwaj *et al.* 1997; Eriksson *et al.* 1997). Such enzymatic treatment is known to preferentially hydrolyze and remove fine-scale fibrils at intermediate levels of treatment (Seo *et al.* 2000). The cited work showed that treatment with cellulase, at a moderate dosage, removed fibrils but did not affect the overall dimensions of the bleached kraft hardwood or softwood fibers. The removal of fibrils from the fiber surfaces was further supported by observations of enhanced drainage rates after the refined fibers had been treated with cellulase. Taken together, the research findings suggest that external fibrillation alone may not be the dominant contributor to strength under all conditions. In the case of softwood fibers, Seo *et al.* (2000) found that cellulase treatment of refined fibers actually resulted in an increase in the tensile strength of the resulting paper, though the opposite effect was observed in the case of hardwood fibers.

Evidence supporting the primacy of internal fiber modifications comes from the work of Hartman (1985), who reported a strong correlation between internal delamination of kraft fibers (*i.e.* "internal fibrillation") and paper strength. In that study, internal delamination was induced by multiple compression cycles of wet paper handsheets, a process that enhanced fiber conformability without generating significant external fibrillation. No corresponding correlation was found between external fibrillation and paper strength. Such findings are consistent with a well-known correlation between paper's density and the development of inter-fiber bonding. It is worth noting, however, that

Hartman's research considered onto within-plane paper properties, not including the Scott internal bond test, for instance. Thus, there is a need for future work that can include that aspect, along with evaluation of the contributions of external and internal fibrillation to bonding.

Similarly, Lundberg and de Ruvo (1978) minimized external fibrillation by carrying out refining at high pulp consistencies (filterable solids levels); the results likewise showed strong increases in paper strength. These gains were attributed primarily to increased internal delamination and increased conformability of the wet fibers rather than to surface fibril development. Page (1989), likewise, did not find a correlation between external fibrillation and paper strength, reinforcing the view that external fibrils are not universally required for strength enhancement. In addition, Kang and Paulapuro (2006) observed a high correlation between paper strength and the fiber saturation point of the kraft fibers refined in highly contrasting ways. Correlations between paper strength and external fibrillation were good only when focusing on one type of refining (*e.g.* friction grinding or Hollander beating) at a time.

Despite these findings, external fibrillation remains readily observable and has been shown to influence certain paper properties under specific conditions. Fibrils at the surfaces of refined kraft fibers can be observed in optical micrographs, especially when using phase contrast illumination (Olsson *et al.* 2001). The cited study, which focused on mechanical pulps, concluded that effects on paper properties due to external fibrillation were less important than other fiber attributes, such as coarseness and conformability, in determining paper properties. However, Wang *et al.* (2007) reported that the Scott internal bond strength of paper, a property evaluating resistance to delamination, rises strongly with increasing fibrillation of kraft fibers resulting from mechanical refining. Such effects were enhanced when friction grinding was employed. That type of refining was shown to preferentially increase the degree of external fibrillation in comparison to other effects of refining, such as increases in conformability. These findings suggest that the contribution of external fibrillation to paper strength is not universal but depends on the specific strength property being evaluated, the refining method employed, and the relative importance of competing mechanisms such as internal delamination and fiber conformability. Particularly, external fibrillation may play a more pronounced role in properties sensitive to out-of-plane interactions, such as internal bond strength, than in in-plane tensile properties. This context-dependent interpretation helps reconcile seemingly conflicting reports and highlights the need to distinguish between different forms of fibrillation and different modes of mechanical failure when assessing their contributions to paper strength.

Evidence Based on Water Removal and Paper Property Development

Water removal

It is proposed here that many of the interdigitated structures present in a finished sheet of paper or nanopaper will already have been present in the suspension just before formation of the sheet. Such a hypothesis leads to an expectation that various flow events occurring during water removal have the potential to either build upon the structures present in the suspension, to break down some of them, or to orient features of pre-existing interdigitation in various ways.

Going back at least to the work of Jayme (1944), it has been known that the development of strength in paper is strongly correlated with the degree of swelling of the fiber cell walls. This assertion has been supported extensively through measurements of the water retention value (WRV), which quantifies how much water remains within fibers after being subjected to sufficient centrifugal acceleration to remove most of the free water from between the fibers and from fiber lumens (TAPPI Useful Method 256; SCAN Method C 62.00; ISO 23714:2007). Studies have established strong correlations between the swelling of kraft fiber cell walls and development of inter-fiber bonding strength (Thode and Ingmanson 1959; Lundberg and de Ruvo 1978; Barzyk *et al.* 1997).

Although the WRV test can definitively show that processes such as mechanical refining increase the amount of water that remains within kraft fibers that are subjected to standard centrifugation, they cannot fully show where that hard-to-remove water had been located. It is traditional to assume that the residual water remaining after centrifugation is within slit-like mesopore spaces in the fiber cell walls (Jayme and Büttel 1968; Stone and Scallan 1968). Some later evidence, however, supports the idea that at least a portion of the retained water after centrifugation may be associated with external fibrillation and fines rather than exclusively with internal cell wall porosity. For instance, Ström and Kunnas (1991) showed that WRV values of refined pulps could be strongly decreased by treatment of refined pulp with cationic polymers. If the effect had been due to the presence of mesopores in the cell walls, then one would have expected relatively low-mass cationic polymers to have had the greatest effects. However, the opposite was true, with high-mass cationic polymers sharply reducing WRV values at low levels of treatment. This behavior was attributed to agglomeration of the fibrillar material and cellulosic fines, which likely facilitated drainage and allowed more water to be removed during centrifugation. These observations are particularly relevant to the present topic, since highly fibrillated material, which may give rise to high WRV values, has the potential to be involved in interdigitation over a range of size levels.

Another way to increase the swelling of cellulosic material is to carry out chemical derivatization, including increasing the number of carboxylic acid groups, for example through carboxymethylation or TEMPO-oxidation (Barzyk *et al.* 1997). The cited article describes how such treatment increases the availability of cellulosic segments at fiber surfaces to interact with each other, leading to stronger bonds. Notably, the cited authors proposed that the increased paper strength could be attributed to inter-diffusion among nano-scale cellulosic features at fiber-fiber interfaces. Such an interpretation aligns with the concept of interdigitation at the nanometer length scale.

Wet web strength

The strength of paper in the wet-web state after sheet formation but before it has been subjected to any evaporative drying provides further insight into mechanisms that operate independently of hydrogen bonding in the dry state. Barnet and Harvey (1980) showed that the wet-web strength of paper, after it has been formed but before it has been subjected to any evaporative drying, is affected by many factors, including the pulp type, the moisture content of the wet web of paper, the proportion of cellulosic fines, and any curled or kinked character of the fibers. With the removal of water from the wet web by vacuum or pressing, the strength of undried paper increases even before significant evaporative drying has occurred (Page 1993). Such increased strength is consistent with the certainty that fibers constituting the sheet become closer together, which can contribute to greater frictional resistance to the sliding of adjacent fibers if the sheet encounters stress. Kibblewhite (1973) proposed such frictional contributions to wet-web strength as an important effect of mechanical refining of kraft pulps.

Building on this concept, de Oliveira *et al.* (2008) interpreted the wet-web strength measurements as evidence supporting “fiber entanglement” as a contributor to overall paper strength. Based on the cited work, interdigitation can be interpreted as a natural extension of mechanical interactions. Fiber-fiber entanglement within the wet web can be expected to mirror temporal and probabilistic fiber adhesion phenomena before fiber sheet consolidation. Interdigitation can be called out when 3D contacts are rendered more frequent by increased surface fibrillation, floc structure, flexibility, and conservation of 3D morphologies. When the hydrodynamic and fiber forming criteria are favorable, the entanglements become persistent, 3D interpenetration. More specifically, interdigitation is thus not merely fiber crossing, but a process-based amplification and conservation of 3D connectivity for structural integration and mechanical performance.

The analysis of de Oliveira *et al.* (2008) addressed the longstanding question of why paper can be strong enough to withstand the tension forces during handling of the paper web even before sufficient water has been removed by evaporation for the development of direct hydrogen bonding between the fibers. It has been well established that capillary forces, acting in combination with friction at fiber-fiber contact surfaces, play a major role in wet-web strength (Campbell 1934, 1959; Page 1993). Persuasive evidence of the effectiveness of such forces can be found in the way that refined kraft fibers tend to establish optical contact over relatively large portions of the facing surfaces of fibers shortly after sheet formation (Page 1960; Robinson 1980). Additional structural evidence includes the pronounced "crimping" observed at fiber crossing points, particularly where fibers intersect at approximately 90 degrees (Page 1985; Hansson *et al.* 1989; Nanko *et al.* 1989).

Because fibers shrink far more in the cross direction (transverse) than in the lengthwise direction when they dry, the crimping process results in effective shortening of many of the fibers in paper, giving rise to a general shrinkage within the plane of the paper. In commercial papermaking, such shrinkage is constrained by applied tension in the machine direction and by frictional constraint in the cross-direction arising from contact between the paper web and dryer fabric surfaces. de Oliveira *et al.* (2008) further noted that the equations predicting very strong capillary forces lose validity once the fibers have achieved close surface contact. Under such conditions, residual capillary forces are expected to be lower than those estimated by Page (1993), suggesting that frictional interactions and mechanical interlocking including potential interdigitation may play a more prominent role in sustaining wet-web strength at advanced stages of consolidation.

Expected effects of stretching web paper or nanopaper

During the mechanized production of ordinary paper, the wet web becomes progressively stretched in the machine direction during the course of wet-pressing and drying. Papermakers use such stretching as a way to avoid the development of wrinkles, and in addition, it helps to develop somewhat higher strength in the machine direction. A similar effect occurs during drying of paper, especially when tension is applied to prevent overall shrinkage. According to Vainio and Paulapuro (2007), at least some of the increased strength of paper that is dried under tension can be attributed to straightening out of fiber segments within the hydrogen bonded structure of a paper sheet. Though the cited work did not consider interdigitated structures, there appear to be several ways in which the concept of activation can be applied. These are briefly summarized in Table 1. Note that the expected effect listed in Table 1, though consistent with the pictorial definitions of the different features of interdigitation, are in need of experimental verification.

Table 1. Some Expected Effects of Minor Stretching on Different Features of Interdigitation

Interdigitation feature	Expected effects
Side-to-side (Fig. 1)	Straightening of the feature, followed by partial slippage
Over-&-under (Fig. 1)	Tightening of the feature, followed by partial slippage
Spanning layers (Fig. 4B)	Tightening of the feature, followed by partial slippage
Entanglement (Fig. 10)	Stretching of network and possible slipping of entanglement nodes
Twisting (Fig. 10)	Straightening of the feature, followed by partial slippage

Evidence of easy separation of paper sheets after pressing and drying in contact

It has long been known that freshly formed, still-wet paper sheets appear to retain a memory of their separate identities, even when stacked, squeezed tightly, and then dried as a group (Hunter 1947). Despite prolonged contact under pressure, the sheets can later be separated with relatively little force, indicating that bonding across the sheet-sheet interface is substantially weaker than bonding formed within each individual sheet during formation. This observation provides early and compelling evidence that fiber interdigitation and the resulting bond development occurs predominantly during the initial sheet-forming stage, rather than during subsequent pressing and drying. This characteristic underpinned traditional papermaking practices in England and continental Europe before the emergence of mechanized papermaking (Hubbe and Bowden 2009). Hand papermakers routinely squeezed posts (meaning stack of several dozen) of wet paper sheets in a screw press and then hung groups of about ten sheets in attic spaces to dry. The fact that these sheets could later be separated without major damage further supports the notion that only limited fiber interpenetration occurs between independently formed sheets even under conditions of high compressive stress.

Yang *et al.* (2019) reported findings in which relatively poor bonding was obtained when two separately-formed layers of TEMPO-oxidized NFC had been pressed together in a wet condition. The layers were readily delaminated and peeled apart after drying. They found that treatment of the interfacial region with a cationic polymer greatly increased the bonding strength. However, the initially poor resistance to delamination is consistent with an assumption that the initial formation of the NFC nanopaper plies took advantage of a high level of interdigitation, and that such interdigitation was absent in the zone between the two pressed-together plies.

Further evidence of preferential bonding within separately formed sheets of paper is apparent in the manufacture of multi-ply paperboard from plies prepared in a system of several separate cylinder formers (Attwood 1991). In such structures, insufficient bonding between adjacent plies, commonly referred to as weak "ply-bonding," can compromise mechanical performance. In practice, this deficiency is often severe enough to necessitate the application of starch or other adhesives to the ply interfaces before they are pressed together (Price and Hubbe 2021). More recently, nanocellulose has been shown to substantially increase ply-bonding within multi-layer paperboard products, presumably by increasing contact area and promoting mechanical interlocking and hydrogen bonding across the ply interface (Starkey *et al.* 2025). Certain two-ply or multi-ply paperboard products are rejected and repulped if the Scott internal bond test is too low. These observations reinforce the conclusion that fiber interdigitation during initial sheet formation plays a decisive role in determining bond strength and resistance to delamination.

Evidence of disruption of paper structure by excessive "action" during formation

Further evidence regarding the importance of interdigitation of fibers within a paper sheet can be gained by measuring the effects of hydrodynamic disruption of sheet structure during the forming process. It is well known that the visual uniformity of paper produced on Fourdrinier paper machines can be improved by judicious application of hydrodynamic shear. This is accomplished by an interaction among millimeter-scale vortex features within the wet web, together with the action of hydrofoils, which are designed to promote more rapid dewatering of the wet paper (Hubbe 2014a). In twin-wire forming systems, the function of hydrofoils can be substituted by "forming blades" on paper machines. The forming blades impose similar hydrodynamic disturbances while the sheet is being formed between a pair of plastic fabrics (Nordström 2006). The local features on a Fourdrinier type of paper machine can be described as transient "volcanoes" in which portions of the fiber mat becomes partly refluidized. While such action can reduce large-scale flocculation and improve macroscopic uniformity, excessively intense microturbulence induced from such devices has been shown to adversely affect the strength of the resulting paper (Nordström and Norman 1996; Nordström 2006). Norman (1986) documented a case in which reduced hydrofoil action resulted in a more floccy visual appearance of the resulting paper yet resulted in higher burst strength. The cited author proposed that, although the paper had a less uniform appearance on direct viewing, it may have had a "uniform structure on a microscale," characterized by better fiber interdigitation and more effective load transfer at the fiber-fiber bond level. This example highlights a critical trade-off in papermaking: conditions that enhance visual uniformity do not necessarily optimize the microscale structural integrity that contributes to strength.

Effects of drying paper on the bonding within recycled paper

Partly irreversible changes are induced in papermaking fibers, especially kraft pulp fibers, each time that paper is dried (McKenzie 1984; Weise and Paulapuro 1999; Hubbe *et al.* 2003, 2007). One of the most widely recognized consequences of drying is closure of some of the mesopores within the cell walls of fibers (Stone and Scallan 1966). This phenomenon, often referred to as hornification, appears to involve the establishment of areas with a high density of hydrogen bonding within and between the fibers (Hubbe 2014b). Although the formation of hydrogen bonds between cellulosic surfaces generally contributes positively to paper's strength, the same interactions become detrimental upon rewetting during the preparation of recycled paper. Specifically, the loss of swelling ability strongly correlates to a lower fiber conformability and less ability to form strong inter-fiber bonds in recycled paper (Klungness and Caulfield 1982). This observation is important with respect to interdigitation, since the same factors can be expected to render external fibrils and nano-scale cellulosic hairs less available for bonding.

Velcro, a fastening system employing hooks and loops, can serve as an interesting analogy to explain how a sequence of interdigitation, followed by drying of a cellulose-based structure, can lead to a somewhat irreversible bonding process. Velcro was inspired by the hooked seed burs of burdock plants that attach to fibrous surfaces by physical interlocking (Arzt *et al.* 2003; Gao *et al.* 2005). According to this concept, the initial interdigitation is able to take place at relatively low energy, due to the favorable orientation of the hooks. But the drying of a cellulosic structure leads to the formation of a dense array of hydrogen bonding at interfaces. So, just like Velcro, the bonds can be more difficult to take apart than to form after the material had been dried.

Reduced swelling limits the capacity of fibers to deform under pressure and achieve contact during sheet consolidation. Beyond bulk conformability, loss of bonding ability might also be attributed to changes at the fiber surface. Particularly, fibrils or microfibrils that normally protrude from the fibers surface and could contribute to mechanical interlocking and interdigitation may become permanently immobilized during repeated drying cycles. Within the constraint of never-dried or well-hydrated conditions, these protruding elements are partially plasticized by interfacial and bound water layers, which can be expected to act as a molecular-scale lubricant. This "hydration envelop" facilitates reversible sliding, reorientation, and penetration of fibrillar elements during sheet consolidation, allowing interdigitation without hornifying. Presumably if such fibrils or microfibrils at fiber surfaces have become permanently pinned down on the fiber surfaces due to extensive hydrogen bonding during drying, they would not be able to interdigitate during a subsequent recycling of paper. This mechanism provides a complimentary explanation to pore closure, helping to account for the observed decline in bonding efficiency in recycled fibers.

Although the loss of swelling ability, due to the drying of paper, can be partly reversed by mechanical refining (Zhang *et al.* 2004), this remediation comes at a cost. Each application of refining tends to further shorten the fiber, degrade the inherent strength of the fibers and generate additional fines, thereby imposing practical limits on the extent to which refining can compensate for drying-induced damage.

Additional evidence of loss of swelling ability and bonding ability has been reported in the case of cellulosic fines (Laivins and Scallan 1996). Such fines, which can be obtained by the screening of pulp from recycled paper, have been found to yield bulkier, less bonded paper sheets when they are added to the fiber suspension (Joseleau *et al.* 2012). These observations are widely attributed to a loss of conformability of the so-called "dead fines." However, a complimentary interpretation might also be due to an inactivation of microfibrils or fibrils at the fiber surfaces. The idea is that the same drying-induced mechanisms leading to loss of swelling ability may be permanently matting down the surface fibrils or microfibrils and preventing them from later becoming involved in interdigitation when forming the next generation of paper. As with fibers, mechanical refining of cellulosic fines from kraft fibers has been found to restore much of the swelling ability (Laivins and Scallan 1996), supporting the idea that structural rearrangements at the fibrillar level are central to their behavior.

Calendering vs. strength

The last unit operation in a typical paper machine is calendering, during which the sheet passes through one or more nips between smooth rolls. The hypothesis here is that though calendering can be expected to break some local structures of connections within a paper-like structure, it is unlikely to contribute to interdigitation. Thus, an important question to consider is whether and to what extent features of interdigitation may be lost, in various cases, as a result of calendering.

The general purpose of calendering is to achieve a smoother surface and uniformity, with the paper also becoming thinner. In addition to those intended changes, it has also been shown that calendering often leads to a measurable loss in paper strength (Burnett and l'Anson 2003). The likely explanation lies in the dry condition of the hot sheet during calendering. In that state, if bonded areas of the sheet become separated, it is too late for new hydrogen bonds to develop between adjacent fibers. Although these observations do not directly depend on whether or not the features of interdigitation fiber structures are present, the potential role of interdigitation cannot be ruled out. This interpretation reinforces the broader theme that the timing, hydration state, and reversibility of bond and interdigitation formation are critical determinants of paper strength development and retention.

Evidence from Cellulose Biosynthesis

Up to this point, much of the cited evidence related to interdigitation has come from observations related to the fiber scale of observation. But there is reason to expect that related phenomena are continually acting at a nanometer scale, even during the original biosynthesis of cellulose. Thus, broader support for the pervasive and highly organized development of high levels of hydrogen bonding within cellulosic materials is evident from studies of cellulose's biosynthesis and regeneration (Ilyin *et al.* 2018; Hubbe *et al.* 2023). In nature, cellulose biosynthesis occurs through an extrusion process that occurs within organelles at the cell walls of plants (Haigler *et al.* 1980; Matthysse *et al.* 1995). As illustrated in Fig. 14, groups of about six freshly formed cellulose chains are extruded simultaneously from the nano-sized natural spinnerets at the cell surfaces, and their detailed molecular structure gives rise to a cellulose-I crystal structure of organization. This propensity for self-organization underscores the strength and specificity of hydrogen-bond-driven association in cellulose. Such self-assembly suggests that similar, though less perfectly ordered, interactions may occur when cellulosic surfaces have become re-exposed during pulping, and bleaching, followed by paper formation (Hubbe *et al.* 2023). However, once these surfaces undergo drying, the same self-organizing tendencies may lead to the formation of energetically stable bonding arrangements that resist reversal and swelling upon rewetting. Such an inability to swell again after drying might explain a lower ability to engage in interdigitation after kraft fibers have been dried and resuspended in water.

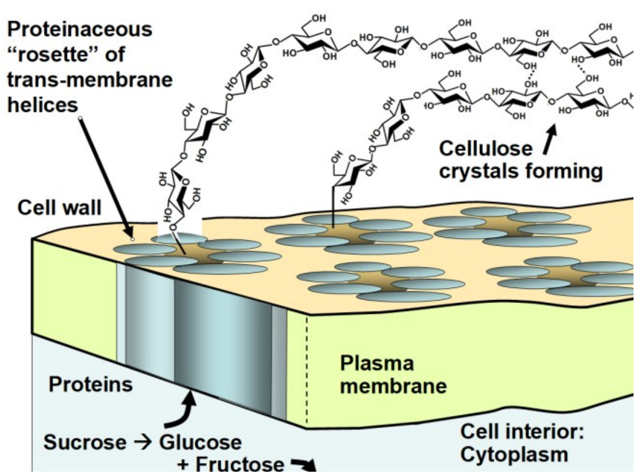

Fig. 14. Depiction of biosynthesis process occurring at a plant cell wall, within which there are rosettes of cellulose synthase, which create glycosidic bonds among sucrose monomers (from within the cell) and extrude individual cellulose chains toward the outside, leading almost immediately to the formation of highly crystalline cellulose nanofibrils

In the case of regeneration of cellulose from solution, analogous evidence has been observed. For instance, it has been proposed that the speed of interdiffusion plays a prominent role in establishing the structure of cellulose films prepared by precipitation of cellulose from M-methylmorpholine (NMMO) solutions (Ilyin *et al.* 2018). Different solvent conditions were shown to lead to different film properties and different types of porosity, highlighting how kinetic factors govern the balance between desirable network formation and excessive consolidation. These findings offer useful parallels for understanding drying and redispersion phenomena in papermaking fibers and nanofibrillated cellulose (NFC), where uncontrolled drying can similarly compromise re-dispersibility and bonding performance.

Evidence Related to Nanocellulose Usage in Papermaking

Importance of aspect ratio

Before considering experimental findings related to nanocellulose, it is important to consider one way in which typical nanofibrillated cellulose (NFC) differs from conventional papermaking fibers. In addition to being much smaller (*e.g.* widths in the range 5 to 50 nm rather than 15 to 50 μm), the NFC also typically has a much higher aspect ratio. The contrast was illustrated schematically in Fig. 10, shown earlier. It is worth noting that within part B of that figure, the NFC is drawn in such a way as to possibly represent TEMPO-oxidized NFC, which is known for its tendency to form well-separated and very thin individual fibrils in the course of moderate mechanical action (Saito *et al.* 2006).

Earlier in this article, when discussing experimental results related to delamination, it was noted that strikingly similar observations have been reported when comparing findings involving a wide range of sizes of cellulosic materials. On the one hand, it was noted earlier that separately-formed wet sheets of handmade paper are known to be readily separable even after press-dewatering and drying of multiple sheets (Hunter 1947). Very similar results were reported by Yang *et al.* (2019) in the case of sheets entirely composed of TEMPO-oxidized NFC. Thus, from the standpoint of delamination evidence, interdigitation phenomena appear to hold true over a very wide range of size of the fibrillar entities.

Nanopaper properties

Due to the high specific surface area, high conformability, and strong hydrogen bonding ability of nanocellulose, there has been a widespread expectation that adding NFC during the production of ordinary paper would increase its strength properties (de Oliveira *et al.* 2008). The cited authors attributed increases in wet-web strength to the formation of physical entanglements among NFC fibrils and between NFC and papermaking fibers. It was proposed that inter-fiber friction was important to keep the entangled features from sliding apart when subjected to the tensile forces imparted by the paper machine's drive system. Multiple studies have also reported positive effects of NFC (or related highly fibrillated cellulose products) on paper strength properties (Gao *et al.* 2011; Eriksen *et al.* 2008; Taipale *et al.* 2010; Salas *et al.* 2019). However, other investigations have shown that the expected strength benefits were small or entirely absent under otherwise comparable conditions (Taipale *et al.* 2010; Salas *et al.* 2019; Barrios *et al.* 2023). This variability highlights that the strengthening potential of NFC is highly sensitive to its state of dispersion, interaction with other additives, and the hydrodynamic history experienced prior to sheet formation.

It is notable that some of the most positive effects of NFC on paper strength have been observed for systems in which there were no chemical additives to the process before the formation of the sheet (Salas *et al.* 2019). While such simplified systems are useful for isolating fundamental mechanisms, there are two inherent drawbacks of such experimental approaches. First, in the absence of any cationic polymer or inorganic coagulant, such as aluminum sulfate, the efficiency of retaining the NFC during paper formation is likely to be low, raising questions about the efficiency and reproducibility of the observed effects. Second, NFC is well known to strongly decrease the rate of water release from a paper web (Salas *et al.* 2019; Hamm *et al.* 2024), a drawback that can lead to a lower rate of paper production.

Inactivation by cationic polymers and flow

To solve both of the forementioned problems, a series of studies was carried out in which the NFC was first coated with an adsorbed layer of cationic starch (Garland *et al.* 2022; Leib *et al.* 2022). The adsorbed starch caused the NFC to be electrically attracted to the predominantly negative surfaces of the untreated cellulosic fibers. After addition of the treated NFC to the fiber furnish, there was a sequential addition of first cationic acrylamide at a dosage of about 0.1% on a solids basis, followed by colloidal silica at a dosage of either 0.1% or 0.2% (Leib *et al.* 2022). It is well known that an optimized sequential treatment with a cationic polymer followed by colloidal silica can strongly boost the rate of water release from refined kraft fibers (Andersson and Lindgren 1996; Hubbe 2005a,b). Consistent with this understanding, laboratory tests showed that the sequence of additives just described was able to achieve dewatering rates in the presence of NFC that were similar to those achieved with no NFC and no other additives (Leib *et al.* 2022; Barrios *et al.* 2023; Atree *et al.* 2025). From a process standpoint, this result represents a significant achievement. Despite the successful recovery of drainage performance, the resulting paper strength results were disappointing (Barrios *et al.* 2023). In one instance, the paper's tensile strength with the treated NFC, cPAM, and colloidal silica was lower than for the base case with just the default furnish without any of those additives. This outcome suggests that improvements in retention and dewatering can come at the expense of the structural contributions that NFC is otherwise expected to provide.

A possible reason to account for the observed lower strength was proposed (Barrios *et al.* 2023), related to the concept of interdigitation. It was argued that the problem could have been due to an inadequate application of hydrodynamic shear in the laboratory work, and that a higher level of shear could be expected to overcome excessive flocculation (meaning bunching together or entanglement) of the nanocellulose. Follow-up laboratory work with a Wet End Process Simulator (WEPS) showed, however, that even at the highest level of shearing accessible with that device, the strength properties remained low in systems employing NFC treated with cationic starch, added to the furnish, followed by cPAM and colloidal silica, and then intense stirring before sheet formation (Kozel *et al.* 2025). Complementary evidence from suspension studies showed that cationic starch-treated NFC in solution tended to exist as clusters rather than as well-dispersed individual NFC particles (Jones *et al.* 2024). Although some re-dispersion of the clusters was observed after application of the highest level of shearing using the WEPS device, substantial clustering continued to be present in suspensions of cationic starch-treated NFC and related mixtures. These findings suggest that once NFC fibrils become self-associated through polymer-mediated bridging or charge neutralization, their nanoscale mobility and thus their ability to interdigitate with fibers or with one another may be irreversibly compromised.

These observations point toward a form of process-induced inactivation of NFC. While chemical treatments and flow conditions that promote retention and drainage are essential for industrial viability, they may simultaneously suppress the very nanoscale interactions, entanglement, interdigitation, and effective surface bonding that underpin the reinforcing potential of nanocellulose. This trade-off highlights the need for additive strategies and processing conditions that preserve NFC dispersion and accessibility while still meeting practical constraints of papermaking.

Rheological Evidence

Networks or floc structures within cellulose fiber or nanofiber suspensions

In the Introduction it had been hypothesized that structures already formed among cellulosic elements within fiber suspensions can persist, at least to some extent, during formation of paper, maintaining some portion of their arrangements in the prepared sheet. Rheological measurement of fiber or nanofiber suspensions provides indirect but informative evidence of transient or persistent structures involving contacts among the suspended particles or fibers (Hubbe *et al.* 2017; Li *et al.* 2020). In particular, the apparent viscosity of the suspension is highly sensitive to the development of continuous or semi-continuous networks among suspended solids.

Figure 15 shows replotted data from Hubbe *et al.* (2017) in which only results for NFC are considered this time. Each red circle in the plot represents a reported result from an extensive collection of published studies involving NFC suspension in aqueous solutions. As shown, the data did not fall onto a single line. Rather, most of the data were found to fall within two empirical lines, both corresponding to an exponent of 5/2 in the log-log plot. The upper plotted line was interpreted to represent systems in which there was a fully contiguous network structure among the nanocellulose particles, whereas the lower (dashed) plotted line was interpreted as representing systems consisting of mainly broken flocs separately suspended in the water. The distance between the parallel lines corresponds to a factor of about 500 in the magnitude of measured viscosity coefficients. These data show that although the presence of NFC makes it possible to achieve very high contributions to suspension viscosity under some conditions of treatment, whether such levels will be achieved in a given situation will depend on many factors, and that disruption of a chemically induced network structure can be expected to give much lower viscosity.

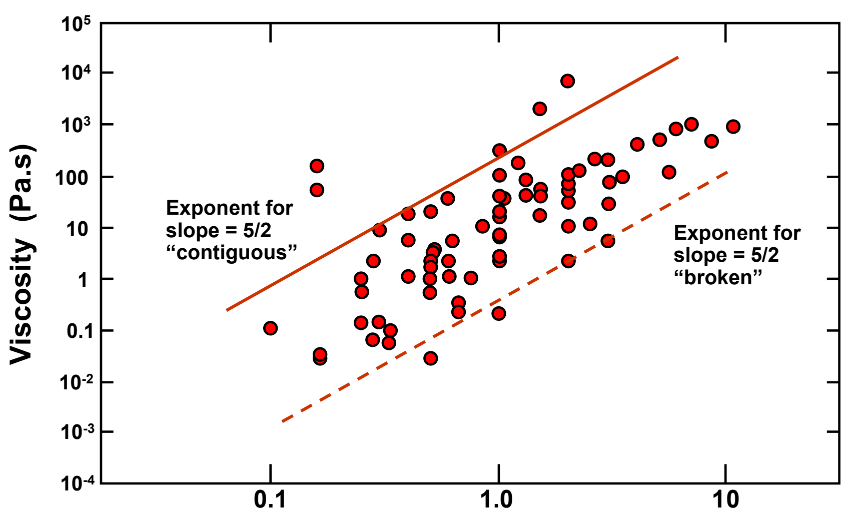

Fig. 15. Replotted data from a review article, featuring only results from rheological testing of suspensions of nanofibrillated cellulose in aqueous suspensions. The plotted lines have the same slope as was reported for cellulose nanocrystal (CNC) suspensions in the original publication (Hubbe *et al.* 2017)

When such networks are disrupted into disconnected fragments by shear, the apparent coefficient of viscosity can fall nearly to the value corresponding to that of the suspending media (Björkman 2003; Hubbe *et al.* 2017). These transitions are often reversible and shear-dependent, consistent with the formation of transient structures rather than permanent aggregates. Direct visual evidence supporting such interpretations was provided by Björkman (2003), who used high-resolution flash photography to observe papermaking fiber suspensions flowing through converging and diverging channels. These images revealed contiguous networks undergoing systematically stretching during flow, consistent with plug-flow-dominated regimes rather than homogeneous shear. Periodic narrow gaps in the floc structures were interpreted as sites where tensile elongation exceeded the mechanical tolerance of the network, resulting in localized rupture. These observations lend physical credibility to rheological signatures of network formation and breakdown, linking macroscopic flow resistance to mesoscopic fiber organization.

Persistent structures present before the paper forming process

As noted in detail by Deng and Dodson (1994), ordinary papermaking fibers in aqueous suspension tend to become locked into floc structures, even when the consistency (filterable solids level) is as low as 0.5%. The cited authors attributed such effects to entanglement among the main parts of the fibers. The formation of fiber flocs has been attributed to random bending of fibers due to small-scale eddies of flow (Steenberg *et al.* 1963; Kerekes and Schell 1992; Dodson 1996). Following such bending events, subsequent straightening of the fibers, due to inherent elasticity within each fiber, can trap them into structures that are held together by a combination of elastic forces and friction. In principle, the mechanical refining of cellulosic fibers, giving rise to external fibrillation, would be expected to provide more opportunities for mechanical interlocking and entanglement. Consistent with this expectation, Yuan *et al.* (2021) observed increasing viscous resistance to flow with increasing levels of fibrillation. Schmid and Klingenberg (2002) carried out calculations to show the reasonableness of fiber flocculation mechanisms involving friction between contacting cellulosic surfaces.

In suspensions of relatively long fibers, such as cotton, the agglomerated features in suspension can be qualitatively different in comparison to those encountered in ordinary papermaking. For instance, Grzelec *et al.* (2025) observed apparent inter-fiber twisted strands. Such features provided the cited author with evidence of handmade papermaking techniques when bast fibers were being used. Related strand-forming behavior has also been observed in suspensions of very-long-chain polyelectrolytes when they were subjected to eddies of turbulent flow in a papermaking suspension (McNeal *et al.* 2005). Such observations suggest shared physical basis rooted in high aspect ratio and flexibility. In the forming of nonwoven fabrics, excessive agglomeration tendencies of long fibers in suspension can be minimized by addition of mucilaginous polymers to the aqueous medium (Hubbe and Koukoulas 2016). A key difference, in the case of very high aspect-ratio fibers, is that the inherent elastic forces within such fibers are much too weak to bring about overall straightening. Rather, effects related to entanglement generally become more important than aspects related to bending and friction among the surfaces of cellulosic entities having aspect ratios much greater than 100.

Gelling effects due to associations

The exceptionally high viscosity of some nanocellulose suspensions has been attributed to overlapping and entanglement among nanofibrils, thereby providing a transient network structure (Pääkkö *et al.* 2007; Alves *et al.* (2024). In such systems, even weak inter-fibrillar interactions can produce pronounced rheological effects due to the high number density and extreme aspect ratio of the fibrils. Alves *et al.* (2024) showed that higher absolute value of ionic charge at the surfaces of nanocellulose fibrils substantially altered these rheological responses, consistent with electrostatic repulsion modulating the extent and stability of fibrillar associations. Under appropriate conditions, nanocellulose particles were thereby able to perform as powerful thickeners of the solutions. Related behavior was reported by Malnaric *et al.* (2024), who observed significant rheological changes in TEMPO-oxidized NFC suspensions upon addition of plate-like graphene oxide particles, suggesting synergistic network formation involving mixed particle geometries.

Strong changes in rheology, which can be attributed to structure-formation among suspended solids, have been reported following the addition of coagulating ions to cellulosic suspensions (Agoda-Tandjawa *et al.* 2012). For instance, gelling effects have been observed upon addition of calcium ions to suspensions of highly fibrillated cellulose or nanocellulose (Agoda-Tandjawa *et al.* 2012; Jowkarderis and van de Ven 2015; Qu *et al.* 2021). Qu *et al.* (2021) showed that the addition of calcium ions to the nanocellulose strongly decreased the absolute value of the negative zeta potential, which is consistent with an interaction dominated by electrostatic effects. Malnaric *et al.* (2024) further reported that addition of calcium ions tended to induce elastic behavior to the suspended material, indicating the formation of mechanically strong networks.

Similarly, stiff gelling effects can be induced by adding cationic polyelectrolytes to nanocellulose suspensions (Jowkarderis and van de Ven 2015), although strong differences in rheological effects have been found depending on which cationic polyelectrolyte was used (Gong 2014). Karppinen *et al.* (2011) observed strong gelation when using a cationic polyelectrolyte to such a suspension, whereas the resulting gel structure was weakened upon adding an amphoteric polyelectrolyte (having both positive and negative charged groups. Vesterinen *et al.* (2010) likewise reported transformation to a solid-like gel state upon addition of cationic starch to a suspension of microfibrillated cellulose.

These findings illustrate that rheology provides compelling, though indirect, evidence for the formation, modification, and breakdown of networks in cellulose-based suspensions. Importantly, while such measurements cannot by themselves confirm the persistence of these structures into the final paper sheet, they strongly support the premise that suspension phase associations whether flocs, strands, or gels create structures that can influence subsequent consolidation and bonding during sheet formation.

CONCLUSIONS

The evidence reviewed in this article supports the view that self-assembly and interdigitation are not peripheral phenomena in papermaking, but rather intrinsic aspects of structure formation that can meaningfully influence strength development in both conventional paper and highly fibrillated cellulose-based sheets. While classical models such as the Page equation have been very successful in explaining in-plane mechanical properties, a growing body of observations points to the importance of three-dimensional connectivity that extends across the thickness of the sheet. Resistance to delamination, in particular, emerges as a sensitive indicator of such out-of-plane interactions. A diverse collection of publications, as considered in this review article, makes it clear that evidence of interdigitation has been available in plain view. That evidence seemingly has been waiting to be gathered into this initial review article, focusing on the concepts and various mostly indirect and qualitative evidence to support those concepts.

Processing conditions during sheet formation including flow fields, flocculation, and drying, *etc*., appear to play a decisive role in establishing opportunities for interdigitation. Hydrodynamic disturbances, partial retention of floc structure, and fiber conformability all contribute to the likelihood that fibers, fibrils, or nano-scale protrusions span across layers and resist separation. Although direct visualization of these structures remains challenging, indirect evidence from fracture surfaces, wet-web strength development, rheological behavior, and internal bond measurements consistently points toward the presence of mechanical interlocking and entangled features formed prior to full drying. In addition, published work supports several contributing mechanisms leading to out-of-plane orientation of some fibers within machine-made sheets of paper. These include the usual overlapping of fibers during filtration of the paper web on forming fabrics, effects of shear flow due to jet-to-forming fabric speed differences, and effects related to the presence of fiber flocs.

The reviewed studies suggest that timing, hydration state, and reversibility of interdigitation and bond formation are critical determinants of final sheet properties. These insights complement established theories of paper strength, by offering a framework to understand strength contributions that arise from three-dimensional organization and transient structures formed during consolidation. Future progress will depend on improved experimental tools capable of resolving out-of-plane connectivity, as well as deliberate processing strategies designed to promote or preserve interdigitated structures. Such advances hold promises not only for conventional paper grades but also for emerging cellulose-based materials where highly fibrillated networks and controlled architectures are central to performance. The indirect nature of much of the evidence can be expected to remain a challenge facing researchers interested in interdigitation phenomena. This challenge is likely to be met in the future by a variety of approaches, including both experimental and based on simulations. In addition, various future product developments based on forming structures from cellulosic fibers and nanocellulose can be expected to benefit from a more complete understanding of interdigitation, in its various forms.

The present review article has emphasized the concepts of interdigitation, as well as various evidence to the occurrence of interdigitation in the formation of sheets from cellulosic fibers or nanofibers. Companion articles, to be published separately, will deal more thoroughly with theoretical issues, including publications shedding light on what can happen at a molecular scale when cellulosic surfaces come into contact. In addition, effects related to the drying of cellulosic materials, from the perspective of interdigitation, will be addressed in a planned review article. A further review article will focus on technological applications that are likely to benefit from advances in this area of science.

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