The metarheology of crowded fibre suspensions

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ABSTRACT

The characteristics of fibre flow are summarised. Major influences on fibre flow research is found to come from paper technology, physics, hydrodynamics, colloidal and macromolecular systems. For a general explanation of technical fibre flow systems, a thermodynamic approach, however, seems more adequate.

INTRODUCTION

Crowded fibre suspensions are common in nature and technology. Skill in processing them is crucial for the outcome of many economically important processes, e.g. in pulp and paper technology, in biotechnology, in production of fibre-reinforced composites, etc. Depending on the fibre concentration, the suspension consistency appears over a wide range from watery, over porridge-like to clod-like.

The problems associated with rheology in connection with fibre flow were commented on in a branch journal by Duffy.[1] A complementary comment to this was submitted.[2] The first part of this contained a summary of fundamentals of fibre flow that cannot be ignored. The second part contained an investigation of the historic background of fibre flow research.

FUNDAMENTALS

i. The uniqueness of fibre suspensions

Paper was as late as in the 1940's by paper technologists regarded as a unique totally stiff material, but is no longer so.[3] Similarly, it is difficult to view fibre flow systems as unique, strange or separated from everyday experience. At low fibre content their behaviour approaches that of the suspending medium (viscous) and at high fibre content that of the fibre material (elastic). At intermediate concentration they display properties of both components.

ii. Material heterogeneity

On atomic level all materials are heterogeneous/particulate. Continuum theory, including microhydrodynamics, cannot therefore be more than one *model* among many possible for fibre flow. A theory is basically just a set of rules to help understanding. A good theory is simple. To describe complex systems with complex models is scientifically less meaningful.

A continuum approach requires enough number of units to be averaged over in a control region. In technical fibre flows, the flow structures (flocs built up by fibres, larger flocs built up of smaller flocs etc., stress chains composed of flocs, whole networks composed of sintered-together flocs and stress chains) at lower and intermediate stress always adapt themselves to the boundaries, which prevents such an averaging procedure. Fibre flow systems are thus heterogeneous/particulate by nature.

iii. Spatial heterogeneity

This heterogeneity, does not just concerns the suspensions, but also the region they occupy. In technical connections, flowing fibre suspensions are enclosed within boundaries, e.g. walls and/or free surfaces.

These constitute prepared slip planes that do not exist in the bulk. The flow effect of a wall may be described with an *efficient wall-slip,* and rheometric methods exist for evaluating this for simple flow fields, e.g. Mooney's method.[4] For general flow fields, and always in practice also for simple flow fields, this is, however, not possible and is something that must be accepted and coped with.[5] Such wall-effects do not seem to diminish or disappear at higher shear rates,[5] possibly since the roughness of the boundaries normally (intentionally) is much smaller than that of the flow structures, i.e. the flocs.

iv. Metarheology

As a result of this spatial heterogeneity it is in practice impossible to extract a "true" rheology, i.e. an apparatus-independent material property, for fibre flow systems from measurements with rheometers.

At lower and intermediate stress, the fibre flocs thus adapt themselves to the vessel geometry. At higher stress the flow structures gradually depends less on the boundaries and finally attain size and form through self-organisation. Then, a "true" apparatus-independent bulk rheology may be defined. But still the wall-effects remain that makes it in practice impossible to evaluate this.

Fibre flow systems thus display elements of both particulate and continuous system. At best they can be said to possess a *metarheology.*

v. Scaling

In absence of a true rheology, *scaling properties* take up a central position in fibre flow.[6] This certainly represents a retreat, but at least does not make hitherto results obtained with rheometers useless but just that they must be reinterpreted and cannot safely be transferred between congruent instruments of different size. This may also seem to be a serious retreat, but is the reality that has to be dealt with, and enhances the importance of flow-mechanistic understanding, i.e. the micro-rheology.

vi. Compressibility

The compressibility concept of continuum theory does not fit fibre suspensions well. The compressibility of both the suspending medium (for process reasons normally watery) and the fibres are small, and thence the compressibility of the entire suspension. A compressible one-phase substance changes volume with pressure. A gas bubble roughly doubles its volume when it ascends from a depth of 10 metres to the surface. A fibre floc sunk to this depth does not decrease in volume just more or less than the liquid. A gas bubble in a non-viscoelastic shear flow field deforms but retains its volume (approx.). A suspended fibre floc in the same shear flow field decreases in volume with increasing shear rate, and vice versa. This volume change can, besides floc surface modifications, be explained in terms of the Poisson ratio difference between the floc and liquid phases.[6–10] I.e. a negative stress component compresses the suspended floc more than a positive component of equal magnitude would do. One-phase systems and fibre floc systems thus differ principally. The fibre system behaviour is not difficult to understand *per se.* It is a continuum view that makes it difficult.

The incompressibility idealisation is often used in continuum theory; so common that it often seems to have been forgotten that it efficiently excludes physical explanation of stress generation in *any* material. According to the author this concept could equally well be dispensed with since "small compressibility" functions equally well, without excluding a physical explanation of stress generation.

vii. Looseness

Strength is a concept that also becomes problematic if transplanted directly from continuum theory to fibre suspensions. Of their own, the fibre flocs have basically no strength in a continuum sense. A floc isolated in a liquid volume substantially larger than itself merely floats apart, i.e. disintegrates into fibres, upon gently mixing (e.g. by a finger) that is just sufficient large to transport the fibres away from the floc, i.e. the flocs are *non-coherent*.[6] Enclosed between boundaries, the flocs offer resistance primarily because the fibres and flocs stand in the way of each other.

In addition, the network does not possess much of internal strength since small loose-jointed shakings, of the type since long and still is practised in hand paper making, cause the fibres change internal position and even out the network. This effect has also since long, and is still, utilised in paper machines (register rolls, wire shakes, foil lists, etc). This behaviour is not difficult to understand *per se.* Again it is a continuum view, developed for materials with an own material strength, that makes explanation problematic.

viii. <u>Laminar and turbulent</u>

At technically relevant fibre concentrations, the overall whiteness of a paper pulp suspension makes observation of internal motion difficult. The author has designed a number of instruments for fibre flow to overcome these problems. Except for plug flow, he has never observed anything that can be described as laminar in the literal meaning of the word. Once the network plug has been broken down and flocculated flow commenced, a complex motion always sets up with more or less transient flocs unpredictably being pushed between each other. Since the tear strength of such flocs also is small, it is natural to imagine them as temporary gatherings of fibres and flocs, i.e. *dissipative structures.*

The turbulence concept of hydrodynamics has also in the literature been applied for fibre flow. One should, however, be aware of that even the foremost authorities in turbulence debate its meaningfulness.[11] Turbulence often appears as just a lumped rest term for what is not understood. Sometimes it seems to aim at irregularities caused by inertia or all deviations from the flow obtained if the inertial term in the Navier-Stokes equation is neglected. That this equation itself is just an approximation of the particulate atomic reality, where these irregularities must have their origin, has not prevented it from being used as a starting point from the start (Reynolds 1887). Nor does the normally used incompressibility assumption seem to be questioned. That such a statistic measure for such complex flow situations should obey other simple laws than the statistical themselves (and the unescapable thermodynamic laws) is difficult to understand. It is another matter when just few and/or well-separated mechanisms are involved as in gas dynamic, atomic physics, etc.

For fibre flow, these irregularities may be of an inertial, viscous, plastic and/or elastic origin. The only definite knowledge is that at very low speed only elastic deformations forces play a role, and when deformation increases, plastic effects caused by fibre/fibre contact point sliding also enter the picture. When flocculated fibre flow starts, viscous and, at even higher absolute speeds, inertial forces also come into play. Whether the elastic and plastic effects ever cease to be significant is not known. Possibly, just their relative importance diminishes. Scaling studies may here eventually throw light. In practice it is, however, difficult to separate the inertial type of irregular motion from the other irregularities. And if it would be possible, then the difficult question remains, which reference should be used? Merely the absence of inertial effects and/or also when the other effects are eliminated? In the first case the reference value cannot be determined, and in the second case the fibres must also be absent, and the reference is then no longer a fibre suspension. If fibre flow regimes anyhow should be labelled, "non-inertial" and "inertial", do at least not say too much.

The introduction of turbulence made the later introduction of *turbulence damping* almost compulsory (Tom 1948). For polymers, this effect was for decades believed to be localised to the turbulent boundary layer close to the tube walls. Some years ago it was, however, demonstrated that this effect starts already upstream from where the turbulence damping polymer added at the tube axis could have reached this layer.[12] The established explanation thus was, at best, incomplete.

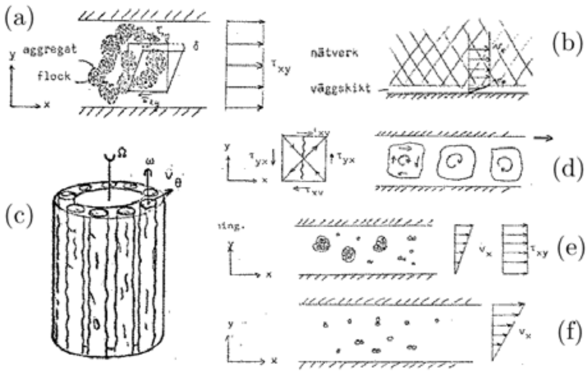
Fibre flows are off and on also discussed in turbulence damping terms but, to the author, then appears to rather be a physical artefact caused by less meaningful comparisons between the fibre suspension and its suspending medium at commom kinematic measures, e.g. volumetric average flow rate, rotational speed, etc.[6] The best way to avoid *extra problems* in fibre flow is probably to avoid the turbulence concept and focus on flow mechanisms.

FIBRE FLOW RESEARCH HISTORY

A historic investigation of background and early development of fibre flow research was undertaken, with the ambition to find the reasons for peculiarities in the direction of present fibre flow research. A number of influences was found; technical, hydrodynamic, physical, colloidal and macromolecular. Their action and interaction, sometimes in one and the same person, has generally resulted in a fibre-centred microhydrodynamic approach. A fuller account of this, partly dramatic, history is given elsewhere.[2,13]

FIBRE FLOW MECHANISMS

The structural development in flowing technical fibre suspensions, *Fig. 1,* and its



Figur 1. Mycelial fibre flow structures in a rotational viscometer (Haake RV2, gap width 2.6 mm, transparent cup).[14] (a) The hierarchial structure of the suspended fibre networks with fibres, flocs, aggregates and floc chains. (b) Plug flow, fibre-free wall layers. (c) Initial network plug break-up. (d) Stress/strain analysis of the primary flocs. (e) & (f) With increasing shear rate, floc size reduction through successive floc splittings.

connection with specific rheological effects, *Fig. 2,* have been known since long[14] although understanding came gradually. This was partly due to difficulties in observing the flowing microbial fibres by eye or in microscope in flow cells (too small focal depth, at that time less developed video technique, etc.) that made a change to the directly observable fibres in wood fibre suspensions necessary. Other at that time noticed but not understood effects included the connection between the break-up and anti-thixotropy and *vice versa,* that no simple rules between fibre parameters (fibre concentration, fibre form, fibre aspect ratio, branchiness, &c.) and rheology existed, that surface chemistry (indicated by fibre wetness, foaming, filtration and sedimentation characteristics, etc.) could easily outweigh the influence of the fibre parameters, the occurrence of "turbulence damping" in pipes and fermenters (i.e. mechanically stirred tanks), etc.[14–16]

Early it was also realised that the flow of these suspensions could be easier understood through a limited number of rules if a floc view was adopted, together with the

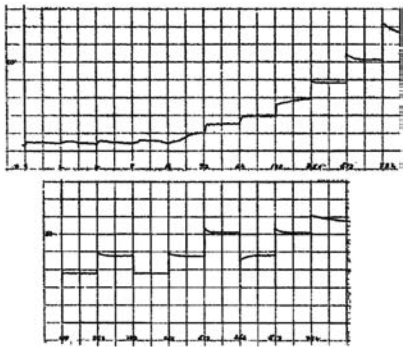


Figure 2. Viscometer torque-time charts for mycelial fibre suspensions displaying; rest stress, characteristic turn from anti-thixotropy to thixotropy, at higher speeds irreversible rheological changes, etc.[14]

the difference in compressibility between the fibre flocs, *Fig. 3*, and the inherent tendency of compressive floc chain formation in largest compression direction. The internal topology of these loosely packed systems consisting of flocs surrounded by similar flocs in all directions namely means that a floc pushed forward in one direction has more neighbours in the forward/sideways direction than in the forward/forward direction, and that the collective focusing action of these initially outweighs the deviating effect of the forward/forward located floc(s).[6,7,10]

i. <u>A standard scenario. Fig. 4</u>

With these results and rules standard scenarios like in *Fig. 4* may be constructed, which in absence of a "true" rheology may

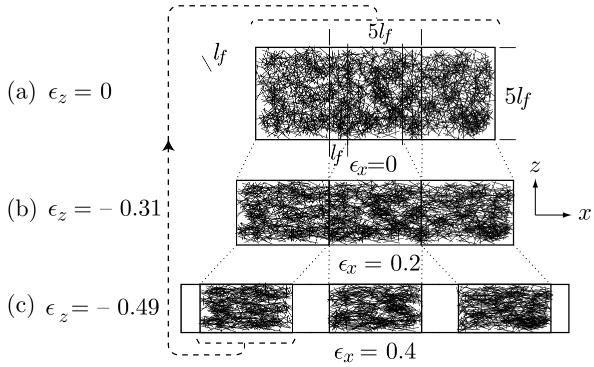


Figure 3. 3-D uniaxial squeezing apart (crushing) of a network into flocs. (a) Three surface-intertwined cubic suspended fibre flocs, so-called *modules.* (b) Compression just until separation. (c) Somewhat further. From Björkman.[6]

serve as practical complement to the scaling properties. This one incorporates most phenomena that have been observed, but does not mean that all phenomena always occur in a specific case. Generally, suspended fibre networks and fibre flow cannot be treated independently. If a fibre flow is stopped, a flocky network forms through a successive sintering-together of larger flocs during the retardation. The non-deformed plug in (a) may be more or less stress-free depending of how completely the flow energy dissipates during the stop. The remainder is frozen-in as elastic energy, stored as an internal isotropic over-pressure. At slightly higher fibre content, this pressure may be large

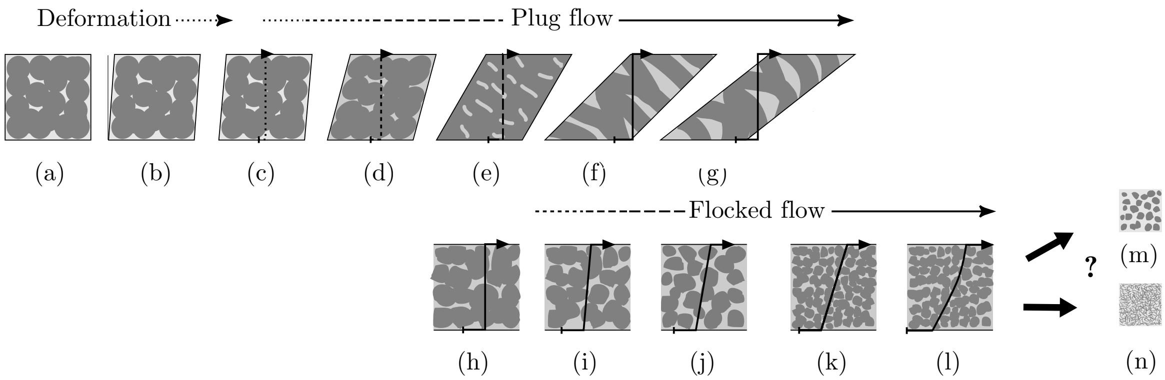


Figure 4. A standard scenario for fibre flow. (a) Undeformed network. (b) Elastically deformed network. (c) Plastically deformed network. (d) Beginning development of compressive floc chains in largest compression direction. (e) Sideways squeezing out of liquid from floc chains. (f) Further lengthwise compression of floc chains. (g) Buckling of floc chains. (h) Separation of floc chains into flocs. (i) Flocculated flow. (j) Floc shrinkage. (k) Floc splitting. (l) Inertial effects. (m) End state asymptotically compressed flocs. (n) End state fully dispersed flocs, i.e. individual fibre flow.

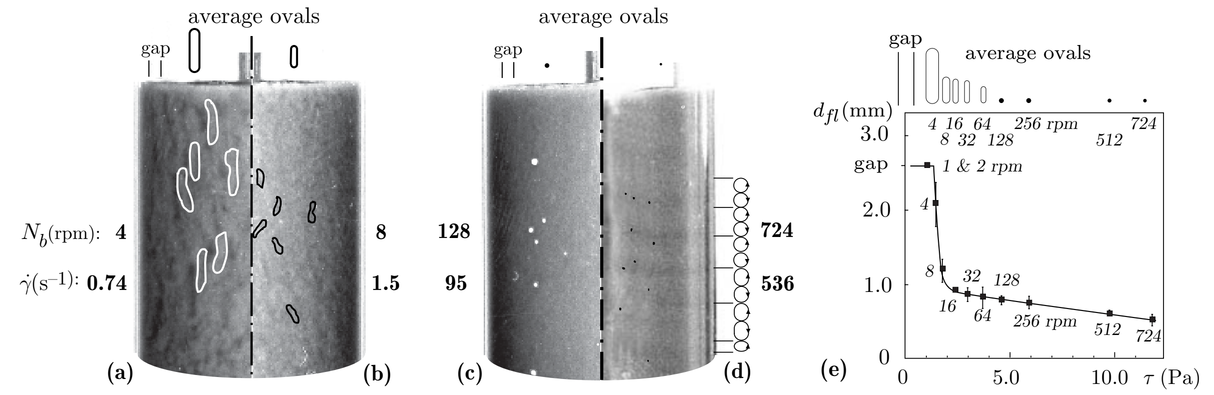


Figure 5. The visual appearance of a flowing 3.1% mycelial fiber suspension, (a) to (d).[5] Average flocs as ovals over free surface. (e) Steady state average floc sizes for the same experiment. Average floc diameter d_{fl}, with standard deviations, *vs.* volumetric average shear stress τ. Fibre average length 0.5 mm.

enough to overcome gravitation, surface tension, fibre entanglement, etc. and press up the flocs forming the characteristic uneven free surface of fibre suspensions.[9,10] Plug flow normally starts before plug break-up has commenced, (c) to (f). Exactly when, depends on circumstances and is difficult to predict. Flocculated flow does normally not start evenly. With a stress distribution in the flow equipment, it spreads from the boundary with highest stress.[6,17] If the stress distribution is sufficiently even, or under non-stationary conditions, it may start in the bulk as a narrow shear band, say a few flocs wide, that widens until fully flocculated flow prevails, about from (g) to (i). *Floc sizing,* the name for the ongoing shrinkage in *Fig. 3,* from (h) to (j) and also from (k) to (l) and *floc splitting,* the name for discrete process when the flocs separate in *Fig. 3*, from (j) to (k) may be repeated until floc diameters reach about fibre length, (n). At higher speeds, a ploughing effect can also sometimes be observed, i.e. due to inertia, flocs have not time to escape sideways when approached by another floc, resulting in a number of flocs pushing each other in the flow direction, (l). Whether or not the flocs disperse fully at the highest deformation rates is of principal interest. The author has, however, never at attainable instrument speeds observed anything else than flocculated flow for *crowded* suspensions, i.e. limit alternative (m).

ii. Shear flow. Figs. 5 & 6

In *Fig. 5a* to *d* four photographs out of eleven show the flow structural development in a Couette instrument with increas-

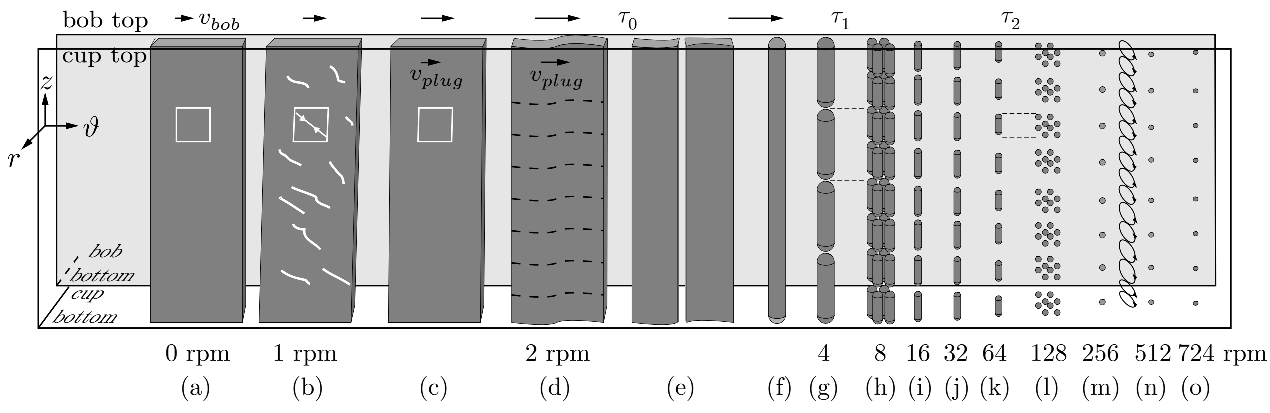


Figure 6. Interpretation of the flow structural observations as exemplified in *Fig. 5.* For explanation see text. Average floc oval dimensions in accordance with *Fig. 5e.* For readability not all flocs, in this always crowded system, are retained after a floc splitting. Taylor vortices start between 256 and 512 rpm.

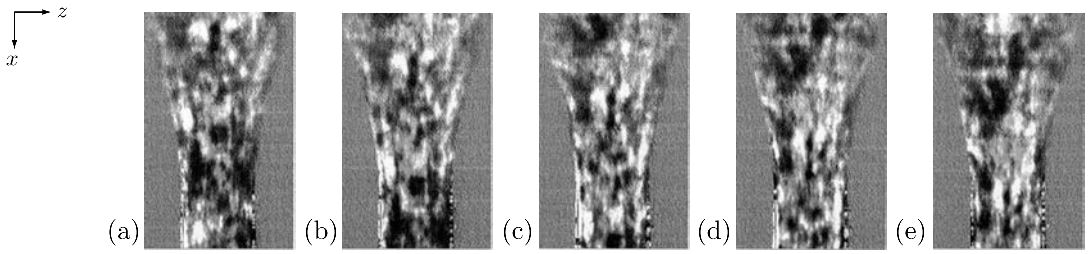


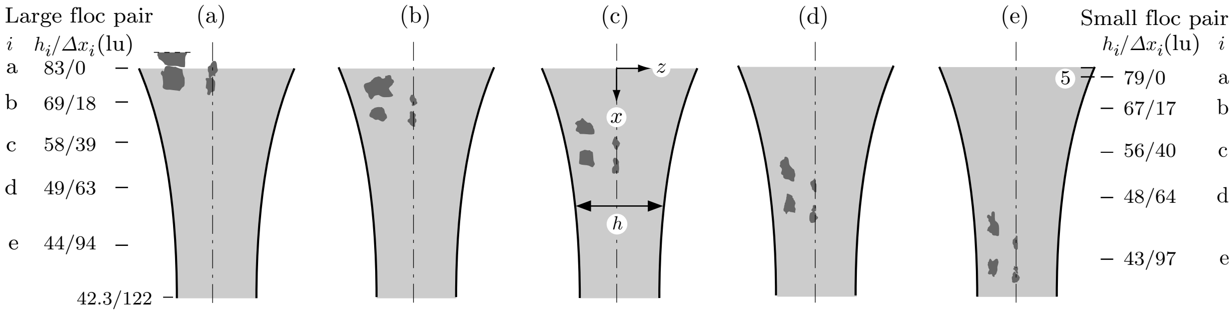
Figure 7. Video frames of suspended wood fibre flocs passing through a contraction.[18,19,21] Time between frames 1 millisecond. The flocs located in the upper section of the first frame have in the last frame reached the lower section. Channel height (in the z-direction) at bottom is 20 mm. The gables are 13 mm apart, i.e. in the y-direction. Fibre concentration is about 0.5% by weight.

ing speed filled with a mycelial fibre suspension. *Fig. 5e* presents graphically the result for the entire series. In *Fig. 6* the same development is interpreted in the terms of the standard scenario.

Initially the stationary plug, through its internal over-pressure, presses against all solid surfaces. The resulting stiction is first overcome at the moving bob and then at the stationary cup. As a result the plug, still attached to the bottom, is sheared horizontally in (a) and (b). When bottom stiction is also overcome, the plug starts to rotate around the axis, (c). The sheet shear buckling in (d) is the first sign of approaching plug break-up in (e), followed by one further axial splitting in (f). Then one axial, one tangential and two radial splittings follow, until in (l) the flocs through self-organisation attain a rounded form. The simultaneous floc shrinkage should also be noticed. The onset of Taylor vortices between (m) and (n) may within the framework of dissipative structures be viewed as *hyperflocs,* i.e. another type of fibre aggregation. The number of fibres in final incoherent round flocs is about 4500.[6]

iii. Contractive flow. Figs. 7 - 10

The headbox of the paper machine consists of a channel contraction that effectuates the transition of the pulp flow from pipe (of, say, 1 m diam.) to a plane jet (of, say, height 2 cm and width 11 m) with a speed of, say, 30 m/s. The author was asked by colleagues to help with explaining the result in *Fig. 7* and (repeatedly) gave his interpretation,[18] but to no avail. In manuscripts for publication they retained the traditional view that flocs are stretched out (borrowed from polymers) and *torn* apart in headboxes,[19,20] claimed the author's theory applied only for higher concentrations than theirs, and added an example to prove their own view. A comment with the following meaning was then submitted.[21]



Figur 8. Floc passage through a contraction.[18] Two floc pairs have manually been traced-out from video frame enlargements of *Fig. 7*. Local channel height h. Floc pair position defined as midway between the narrowest part of the streak between them. The position of a floc pair relative to that of the same floc pair in (a) is designated $\Delta x_{(i)} = x_{(i)} - x_{(a)}$ with $i = \{a, b, c, d, e\}$, in length units (lu) of the graphic program (Adobe Illustrator). The lengths in millimetres obtained through multiplication with 0.475.

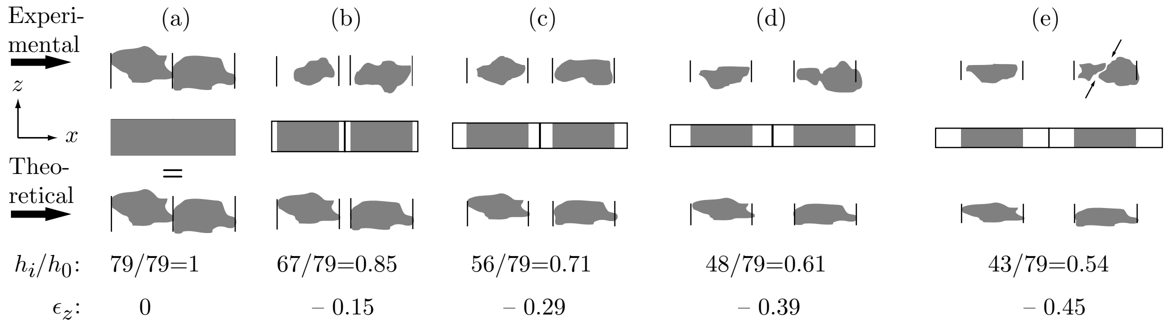
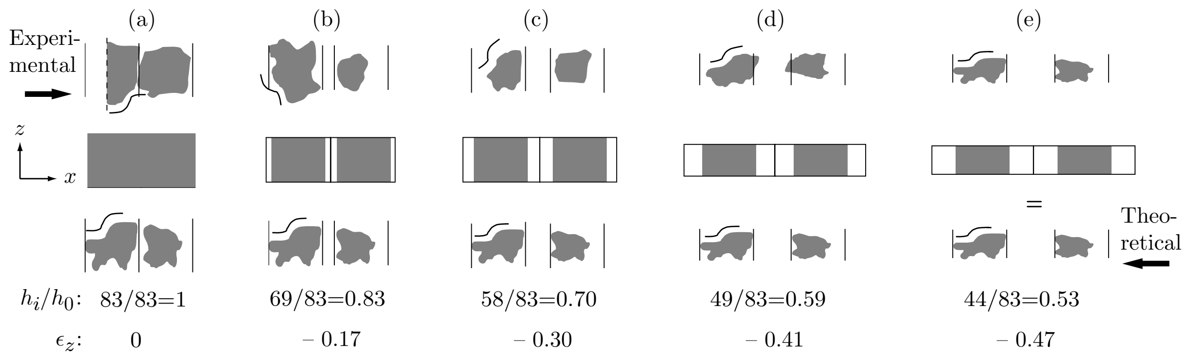


Figure 9. Comparison between experimental and theoretical floc development with a 2-D squeeze-out model.[18] The floc pairs in upper row copied from the on-axis floc pairs in *Fig. 8.* The middle series displays the module (see text) development with Poisson ratio zero for the network and 0.5 for the liquid, c.f. *Fig. 3.* The initial module pair has been drawn to just circumscribe the initial floc pair. The subsequent development is obtained through exact scaling using the same graphic program as was used to draw the rest of the figure. The first floc in the lower series is copied from the first photographed floc pair. The subsequent development is obtained through graphic scaling as for the modules.

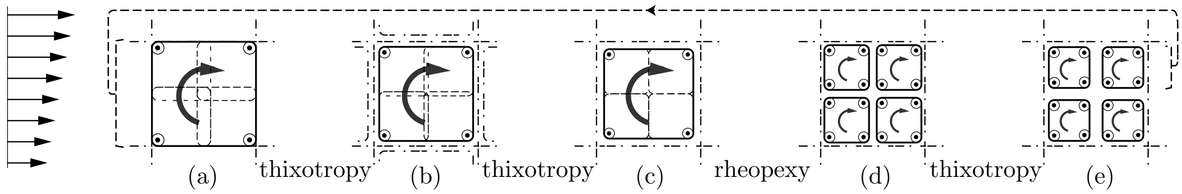
Fig. 8 reveals that the main floc deformation is in the cross direction while the deformation in the flow direction is smaller. This conforms less well to the traditional stretching view. The interpretation in terms of the standard scenario is shown in *Figs. 9 & 10.* Since the contraction is shallow and the gables impermeable a Hele-Shaw approximation is adopted, as is shown in *Fig. 9 & 10,* contrary to in *Fig. 3.* The z-compression is evaluated from the local channel width h_i. Preliminary it is assumed that the squeezing-out is symmetric around the floc pair centres.

A comparison between the upper and lower rows in *Figs. 9 & 10* reveals a good general agreement between experiment and this very simple model, which basically just is phase balances and the Poisson ratio difference. Naturally, not all effects, e.g. the wall-drag-induced rotation of the off-axis floc in *Fig. 10,* can be accounted for, but this may be included with the module theory discussed below. It is difficult to imagine that a fibre-based CFD-model, even with the help of supercomputers, can outperform this, especially since then the natural variations between the fibre can never be fully includes, which in practice are inaccessible to the extent that such a treatment formally requires.

The experimental result in *Fig. 9d* and *e* finally shows crushing of the leading small floc between crosswise strains of -0.39 and -0.45. The theoretical value, based on width of entanglement zone equal to one fibre lenght, is -0.33, compared to -0.31 for the 3-D squeeze-out in *Fig. 3.*



Figue 10. Repetition of *Fig. 9* for the larger off-axis floc pair, with the differens that the lower series is synthesised from right to left beacuse the trailing floc in the initial frame was partly outside the frame.[18]



Figue 11. Example of response of a 2-D module shear flow system upon a step increase in shear rate.

iv. <u>Generalities</u>
In both the shear and contraction examples, a *squeezing apart* mechanism (crushing) due to the difference in compressibility (or Poisson ratio) between the flocs and the liquid may explain the general structural development. Under flocculated flow conditions also viscous surface flattening and eventually fibre ablation from the flocs contribute to the floc size change.[5,6,17]

THE MODULE SUSPENSION THEORY

A *module* is here the name of a rectangular model floc from which enclosed liquid can be squeezed out, *Figs. 3, 9 & 10.* Modules are constituents of the *module suspension theory.* Like real flocs they may also roll, *Fig. 11,* to confirm with shear flow scenarios as in *Fig. 6.* Their translation and rotation is governed by force and torque balances.[5,6] Reasonable agreement between this theory and Couette metarheograms has been achieved.[6]

An advantage with modules is that they facilitate keeping track of the phases. It may objected that real flocs are neither square nor similar in magnitude. This is correct, but the modules are primarily intended as mental aids. The uniform fibres composed of stiff or linked rods used in CFD modelling are neither realistic. Real wood fibres are individual with properties impossible to keep track on. It is good theoretical practice not to found theories on unattainable parameters.

If a fibre flow is successfully modelled as individual fibre flow, and it can be observed that the fibres move in groups/flocs, the model can nevertheless not be more than curve fitting in disguise. The same result can then normally be obtained with less labour with dimensional analysis.

Due to historic reasons,[13] modelling of fibre flow has approached the technically interesting crowded systems from dilute conditions. As a result of this, and hereof inspired comparisons with colloidal systems, these fibre flocs have often come to be regarded as the outcome of a flocculation process when the reality normally is the opposite, namely a *splitulation* process and freezing of dissipative flow structures.

The main purpose with the module theory is scientific, *viz.* to help identifying the basic flow mechanisms and in addition find the minimum number of necessary mechanisms. The theory is based on *observable* mechanisms. In a natural way, it incorporates the metarheological aspects. A by-product may be more realistic micromechanical models that also includes the scaling properties. Basically the theory is, however, thermodynamic and abstract. Its simplicity allows analytical solution of flow problems,[6] which in turn allows testing of thermodynamic least action principles for fibre flow systems.[22]

Fig. 11 gives a 2-D example of the structural response of an enough large step increase in shear rate, say a doubling, to induce floc splitting. The liquid flow in the *streaks* between the modules is Newtonian and laminar. The primary response comes from the boundary streak and results in a doubling of wall stress. The secondary response comes from the modules, which begin squeezing out liquid. This widens the streaks, which decreases wall stress, i.e. *thixotropy* between (a) and (b). At (c) the modules have been compressed to the point that the network cannot keep together. The daughter modules then separate and the liquid is distributed over more streaks, which narrow, resulting in stress increase, i.e. *rheopexy* (anti-thixotropy) between (c) and (d). The system then (with new subsidiary conditions in Euler-Lagrange's equation for dissipation rate) continues its attempts to minimise dissipation through balancing restricted (intra-floc) and unrestricted (inter-floc) deformation. Eventually, the scheme must be iterated a couple of times before a new minimum has been reached. Since these groupings are non-coherent it is natural to interpret them thermodynamically. At higher speed inertia has to be accounted for.[22] Distribution strategies of this type are very general and can applied to almost any liquid (with flocs substituted with atoms and the suspending medium by space itself, and what could be less compressible).

Since chemical/thermodynamic and fluid mechanical influences may be of equal magnitude the choice is to express fluid mechanics in thermodynamic terms or *vice versa.* Since the latter is generally impossible (and would be utterly impractical) the first alternative seems to be natural.

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