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Micromechanical Behavior of Fiber Networks

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Measure what can be measured and make measureable what cannot be measured.
Gallileo Gallilei

Abstract

Paper is used in a wide range of applications, each of which has specific requirements on mechanical and surface properties. The role of paper strength on paper performance is still not well understood. This work addresses the mechanical properties of paper by utilizing fiber network simulation and consists of two parts.

In the first part, we use a three-dimensional model of a network of fibers to describe the fracture process of paper accounting for nonlinearities at the fiber level (material model and geometry) and bond failures. A stress-strain curve of paper in tensile loading is described with the help of the network of dry fibers; the parameters that dominate the shape of this curve are discussed. The evolution of network damage is simulated, the results of which are compared with digital speckle photography experiments on laboratory sheets. It is concluded that the original strain inhomogeneities due to the structure are transferred to the local bond failure dynamics. The effects of different conventional and unconventional bond parameters are analyzed. It has been shown that the number of bonds in paper is important and that the changes in bond strength influence paper mechanical properties significantly.

In the second part, we proposed a constitutive model for a fiber suitable for cyclic loading applications. We based the development of the available literature data and on the detailed finite-element model of pulp fibers. The model provided insights into the effects of various parameters on the mechanical response of the pulp fibers. The study showed that the change in the microfibril orientation upon axial straining is mainly a geometrical effect and is independent of material properties of the fiber as long as the deformations are elastic. Plastic strains accelerate the change in microfibril orientation. The results also showed that the elastic modulus of the fiber has a non-linear dependency on a microfibril angle, with elastic modulus being more sensitive to the change of microfibril angle around small initial values of microfibril angles. These effects were incorporated into a non-linear isotropic hardening plasticity model for beams and tested in a fiber network in cycling loading application model, using the model we estimated the level of strains that fiber segments accumulate at the failure point in a fiber network.

The main goal of this work is to create a tool that would act as a bridge between microscopic characterization of fiber and fiber bonds and the mechanical properties that are important in the papermaking industry. The results of this work provide a fundamental insight on mechanics of paper constituents in tensile as well as cyclic loading. This would eventually lead to a rational choice of raw materials in paper manufacturing and thus utilizing the environment in a balanced way.

Sammanfattning

Papper används i många applikationer som ställer krav på dess mekaniska egenskaper och ytegenskaper. Dock är det fortfarande oklart hur det mekaniska beteendet påverkas av papperets styrka. Fibernätverksmodellering används här för att undersöka pappers mekaniska egenskaper. Arbetet består av två delar.

I den första delen används en tredimensionell fibernätverksmodell för att beskriva brott i papper. Modellen tar hänsyn till olinjäriteter på fibernivå (material modell och geometri) och bindningsbrott. En spänning-töjning kurva av papper i dragbelastning beskrivs med hjälp av det nätverket bestående av torra fibrer; de parametrar som dominerar formen på denna kurva diskuteras. Utvecklingen av nätverksbrott simuleras och jämförts med digital speckle fotografering experiment på labbark. Man drar slutsatsen att de ursprungliga inhomogeniteter i töjningen på grund av strukturen överförs till de lokala bindningsbrott dynamiken. Effekter av olika konventionella och okonventionella bindningsparametrar analyseras. Det har visat sig att antalet bindningar i papper är viktigt och att även små ändringar i bindningsstyrka påverkar pappers mekaniska egenskaper avsevärt.

I den andra delen utvecklas en konstitutiv modell som tar hänsyn till cyklisk belastningshistoria. En fibermodell baserad på tillgängliga litteraturdata och en detaljerade finit element modell av massafibrer föreslås. Modellen används för att visa hur olika parametrar påverkar de mekaniska egenskaper hos massafibrerna. Vid axiell belastning visade det sig att förändringen av mikrofibrillorientering främst var en geometrisk effekt, och därmed var oberoende av materialegenskaperna, så länge deformationen var elastisk. Plastiska töjningar påskyndar förändringen i mikrofibrillorientering. Resultaten visade också att elasticitetsmodulen hos fibern har ett icke-linjär beroende av mikrofibrillvinkeln, samt att elasticitetsmodulen är mer känslig för en förändring av mikrofibrillvinkel vid små initialvärden för mikrofibrillvinkeln. Dessa effekter formuleras som en icke-linjärt isotropt hårdnande plasticitetsmodell för balkar. Modellen testades i ett fibernätverk under cyklisk belastning, där en uppskattning av töjningsnivån på fibersegment vid fibernätverksbrott gjordes.

Målet har varit att skapa ett verktyg som kan ge en förståelse mellan mikroskopiska mätningar på fibrer och bindningar och mekaniska egenskaper av paper. Resultatet ger ett unikt verktyg som kan användas i pappersindustrin för att ge en fördjupad förståelse av pappersmekaniken både under statisk och cykliskt dragbelastning. Detta skulle kunna användas för att bättre optimera råvaran i tillverkningsprocessen, och därmed minska pappersindustrins miljöpåverkan.

Preface

The work presented in this thesis has been carried out at the Department of Solid Mechanics, Royal Institute of Technology (KTH), Stockholm between November 2010 and March 2013 within BiMaC Innovation Research Centre, the financial support of which is sincerely acknowledged.

I would like to thank my supervisor Docent Mikael Nygårds for ideas, comments and an endless stream of articles. My thanks are extended to my second supervisor Associate Professor Artem Kulachenko for guidance and inspiring collaboration as well as for personal advices on how to become a good researcher. I express my deep gratitude to both my supervisors and appreciate the fruitful discussions, support and encouragement from both of them.

I wish to thank my colleagues and friends at KTH Solid Mechanics for being around. I am also thankful to Sylvain Galland for the collaborative work at Wallenberg Wood Science Center, Stockholm, Sweden. I wish to appreciate the help of my friends Prashanth and Kate for the linguistic revision of the manuscript.

Finally, I would like to thank my mom for letting me as a 13-year old to leave home to study what I liked, and to thank my brother and my sister for making me feel special. Last but absolutely not least, I wish to express my sincere feelings to Elias for his love and support through all these years: *ana bähibäk ktir*.

Stockholm, April 2013

Svetlana Borodulina

List of appended papers

Paper A: Stress-strain curve of paper revisited

Borodulina, S., Kulachenko, A., Galland, S., Nygårds, M.
Nordic Pulp and Paper Research Journal, 27(2), 2012,318-328.

Paper B: Constitutive modelling of a paper fibre in cyclic loading applications

Borodulina, S., Kulachenko, A.
Report 541/10. Department of Solid Mechanics, KTH Engineering Sciences, Royal Institute of Technology, Stockholm, Sweden.

In addition to the appended paper, the work has resulted in the following publications:

Stress-strain curve of a fiber network
Borodulina, S., Kulachenko, A., Nygårds, M. *Proceedings of the 23rd International Congress of Theoretical and Applied Mechanics ICTAM*, Beijing, China, August 19-24, 2012.

Influence of paperboard structure and processing conditions on forming of complex paperboard structures
Östlund, M., Borodulina, S., Östlund, S. *Packaging Technology and Science*, 24:331-341, 2011.

3D-forming of double-curved paperboard structures for packaging applications
Östlund, S., Östlund, M., Borodulina, S. *Progress in Paper Physics Seminars* (poster), Graz, Austria, 2011, pp.323-325.

Contribution to the papers

The author’s contributions to the appended papers are as follows:

Paper A: Principal author. Kulachenko’s previously developed fiber network model for wet fibers was further extended to dry fibers by incorporating the appropriate fiber bonding conditions and constitutive relations. It was utilized together with the author. Experimental work has been conducted together with Galland. Both the principal author and Kulachenko contributed to writing process. Nygårds contributed with comments.

Paper B: Principal author, conducted constitutive modeling of a fiber and performed simulation work and results evaluation. Both the principal author and Kulachenko contributed to writing process.

Contents

Paper A

Paper B

Introduction

Papermaking is a multidisciplinary technology with strong competition. The complexity of the processes with up to 100 km/h production speed makes paper one of the fastest continuously produced material and paper-making — one of the most technologically advanced industries. Despite significant advances in paper manufacturing process, it is threatened by the rapid developments in IT-world (such as increasing number of electronic reading devices and e-publications, digital advertisements via Facebook and Twitter, etc. [1] and growing interest for efficient energy usage maintaining economical profit [2]). However, environmental demands on a rational choice and usage of fewer raw materials in production and preparatory testing are a driving force for development.

Paper is a stochastic network of cellulose fibers (Figure 1) with pores in-between filled by other constituents like fines (small fiber fractions), microparticle fillers (clay, calcium carbonate, talc, etc.), various coatings, binders or pigments [3]. Such random networks are found in other materials as well, for example in collagen matrix, carbon nanotubes soot or composite cotton nanotubes. In paper, a network of fibers is held by fiber-to-fiber bonds, which arise during the drying processes in papermaking; the nature of these bonds is still not well explored. The structural inhomogeneity is caused by the sheet formation process [4] that

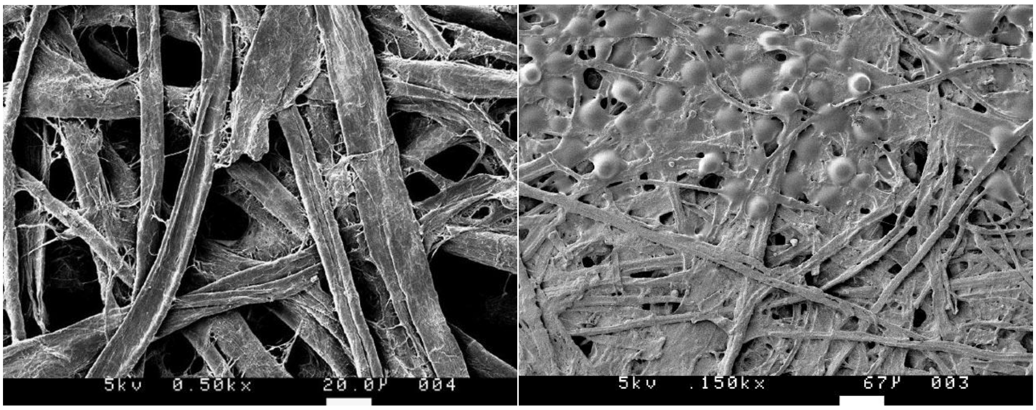

Figure 1 Scanning electron microscope (SEM) image of a paper surface (courtesy of Jim Ekstrom): left – copy paper, right – Post-it paper (partly showing the sticky part with glue particles on the top of the picture).

creates two principal directions called machine- and cross-machine direction (MD and CD, respectively) developing anisotropy. Besides that, this process-dependent disorder is combined with large variations in properties of fibers that originate from different parts of wood; and creates a very complicated, but nevertheless, interesting structure we call paper.

Over the past few decades, structural characterization of paper was performed mostly experimentally and related the properties of paper major constituents (fibers and bonds) to paper sheet properties by variety of methods [5]. With the development of new testing techniques back in 1950-s, it became possible to perform testing on single fibers [6], but the strength of bonds is generally more complicated to measure due to small bonded areas (often referred as relative bonded area (RBA)) and the process of fiber-to-fiber bond manufacturing. It has also been argued about the meaning of the bond strength [7] and the values of it reported [8]. At the same time, the role of disorder on the mechanical properties of paper is indeed abstract and neither cannot be measured nor attributed experimentally. One of the solutions to overcome these difficulties would be to treat paper as a continuum or a fiber-reinforced composite. However, composite approach is unrealistic, as paper does not have a distinct “matrix” and its behavior is much more complex due to the presence of bonds. On the other hand, paper as continuum does not answer the key question whether paper properties are governed by the randomness in fiber distribution, fiber micromechanical ultrastructure, the bond strength, sheet inhomogeneity or a combination of these.

The importance of these parameters in paper could be solved in another context, namely by performing numerical simulations of the network structure, which is also a better alternative in terms of efficiency and performance. Numerical methods in network modeling provide far more possibilities in assessing valuable information, which is not possible to measure experimentally.

Mechanical response of paper sheets to external in-plane tension is usually described by the load-elongation curve, often called a stress-strain curve (SSC). This term is slightly inaccurate because there exists uncertainty of which value to use for the thickness (also called caliper) of a paper sheet as the surface of it is uneven; and also, because stresses and strains are not constant within specimen. Elimination of thickness in the equation for density led to unification of “basic weight” term, which is defined as mass per unit area, usually expressed in units of grams per square meter. We shall nevertheless use the term SSC interchangeably with load-elongation curve.

A typical stress-strain curve of paper from a standard tensile test is approximately linear at small strains with Young’s modulus defining initial slope; also, it has a distinct plastic region prior the failure (Figure 2). When the specimen fails, the end point of SSC defines strain at break and tensile strength. Rheological properties of paper define it to be a visco-elastic plastic material [9].

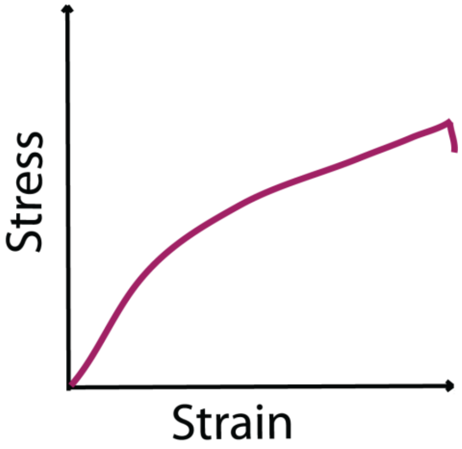

Figure 2 Typical stress-strain curve of paper in tension

Fibers are the principal structural elements of paper [10] and it is recognized that the inelastic features of the hardening behavior of paper is due to the fibers and surprisingly not the bonds [11]. This conclusion was justifiably fascinating as the natural explanation of dissipation during plasticity was attributed to the breakage of fiber-to-fiber bonds.

It comes natural to wonder what controls such inelastic fiber behavior. Let us have a look at fiber architecture in detail. A natural wood fiber consists of several cell-wall layers and has a helical internal structure, as shown in Figure 3.

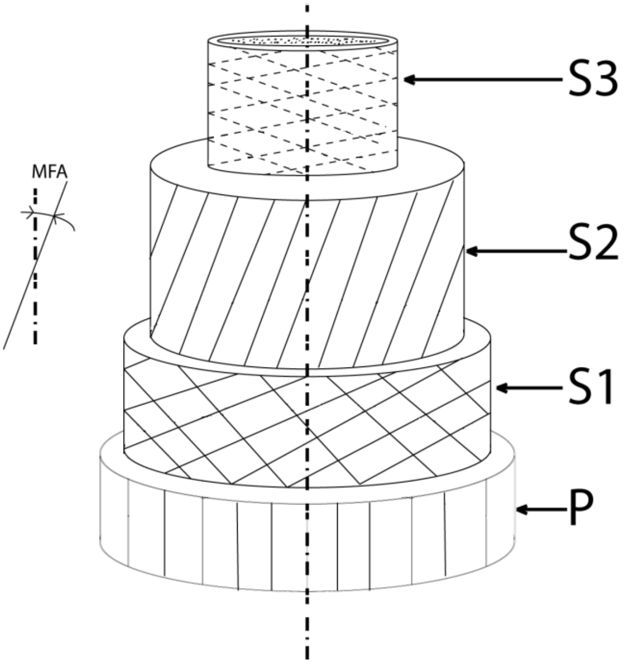

Figure 3 Schematic representation of wood fiber. The following layers are shown: P - primary cell wall, S1, S2 and S3 are secondary cell-walls. The angle of microfibril orientation with respect to fiber axis in S2 is depicted as microfibril angle (MFA). Redrawn from [12]

Cellulose is the most important constituent of the cell-walls and is responsible for load-bearing capacity of the fiber. The thickest of all layers is S2, comprising approximately 70-80% of the fiber volume [13–15]. Fiber's structural stiffness can mainly be characterized by the properties of cellulose microfibrils embedded in a polymeric matrix of S2 layer. The upper estimation for the crystalline stiffness of cellulose is 134 GPa [16], which may be compared with the elastic stiffness of ceramics or engineering alloys.

Ultrastructure of a fiber includes microfibrils that are wound around the axis of the fiber and form a helical assembly in bundles. It is certain that the key feature is the orientation angle of the microfibrils, also called microfibril angle (MFA) measured with respect to the fiber axis. This orientation angle governs the mechanical properties of the fiber ([14], [15], [17–19]). To prepare pulp for papermaking, wood is fractionated and separated into single fibers by means of mechanical or chemical treatments, called pulping processes. This modifies the cell structure and the composition of the cell-walls irreversibly. Thereby, initially damaged fiber after pulping may have another structure, as shown in Figure 4.

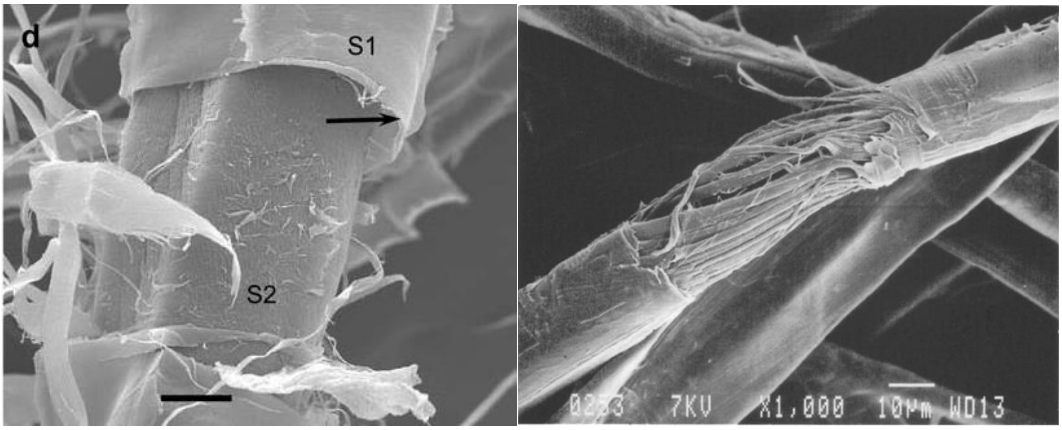

Figure 4 (Left) SEM image of pine thermo-mechanical (TMP) pulp fibre, reproduced with the permission of the authors [20]. Bar 10 µm. (Right) SEM image of a refined pulp showing disruption of the S2 layer due to cyclic loading, reproduced with permission from Springer, Blackie Academic & Professional, ©1995 [21]

Finally, it should be noted that this licentiate thesis includes the combination of *performed experiment* and *numerical simulation* of paper as a network of fibers, as well as *comparison* with experimental data available in the literature. Future work may be concentrated on developing a complex model that accounts for interaction between ribbon-like loosely connected microfibrils as depicted in the figures above.

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Summary of appended papers

Paper A: *Stress-strain curve of paper revisited*

We have investigated a relation between micromechanical processes and the stress-strain curve of a dry fiber network during tensile loading. By using a detailed particle-level simulation tool we investigate, among other things, the impact of “non-traditional” bonding parameters, such as compliance of bonding regions, work of separation and the actual number of effective bonds. This is probably the first three-dimensional model which is capable of simulating the fracture process of paper accounting for nonlinearities at the fiber level and bond failures. The failure behavior of the network considered in the study could be changed significantly by relatively small changes in bond strength, as compared to the scatter in bonding data found in the literature. We have identified that compliance of the bonding regions has a significant impact on network strength. By comparing networks with weak and strong bonds, we concluded that large local strains are the precursors of bond failures and not the other way around.

Paper B: *Constitutive modeling of paper fiber in cyclic loading applications*

This paper investigates the influence of geometrical and material parameters on the mechanical response of the pulp fibers. A three-dimensional finite element model of the fiber is proposed, which accounts for micro-fibril orientation of cellulose fibril and the presence of lignin in the secondary cell wall. This study shows that the change in the microfibril orientation upon axial straining is mainly a geometrical effect and is independent of material properties of the fiber as long as the deformations are elastic, plastic strains accelerate the change in microfibril orientation. The results also showed that the elastic modulus of the fiber has a non-linear dependency on microfibril angle. Based on these findings and supported by available experimental evidences, we propose a non-linear isotropic hardening plasticity model for beams. The model was tested in cyclic load applications in a fiber network. By using the model, we estimated the level of strains that fiber segments accumulate near the failure point in a fiber network.

Errata

Paper A. The dimensions for the contacts stiffness (CS) in both normal and tangent directions should read as ”$1 \cdot 10^{9}$ N/m” instead of “GPa” throughout the paper:

Location	Error
p.320 Table 2	CS should read as ”$1 \cdot 10^{9}$ N/m” instead of “GPa”
p.326 Table 4	CS should read as ”$1 \cdot 10^{9}$ N/m” instead of “GPa”
p.326 Figure 21	Contact compliance should read as ”$1 \cdot 10^{-9}$ m/N” instead of “GPa^{-1}”