New Insights in Stressing Leather – Multi-Scale Simulation of Macroscopic Structure-Property Relations

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ABSTRACT

The naturally grown leather structure is the origin of both the uniqueness of each leather product as well as the inherent variance inside the material. For optimal exploitation of the leather's potential during processing and utilisation, an extensive material characterisation is imperative. However, the qualification of leather properties remains a challenge as conventional testing methods often require the destruction of the test material. In addition, non-destructive computer-aided design and numerical analysis of the physical and mechanical behaviour of components are increasingly essential for industrial mass production. Dedicated software solutions for leather materials are not yet available. As part of ongoing research activities, imaging analysis methods using micro-computed tomography (µCT) were utilised to analyse leather structure areas and corresponding sub-structural elements. The extraction of hierarchical leather structures was facilitated by the use of automated segmentation and parameterisation algorithms. The creation of a stochastic leather structure model was achieved. This pioneering approach enabled the multi-scale simulation of elastic and viscoelastic structural properties, exemplified by the tensile strain testing of leather, using advanced mathematical-numerical simulation techniques. The macroscopic simulation results were validated using classical mechanical and single-fibre tensile tests. These tests showed excellent agreement for the elastic but not for the viscoelastic deformation scales. It is hypothesised that an unexpected loss of anisotropy in the macroscopic leather structure composite occurred during sample preparation for the µCT studies.

Keywords: hierarchical leather structure, micro computed tomography, structure modelling, multi-scale simulation

1 Introduction

Leather is a renewable material that is primarily used in manufacturing of valued consumer goods like shoes, clothing, upholstered furniture, or automotive interiors. Due to the naturally grown raw material from animals' skins, each up-cycled tanned hide for leather production is unique. Being proclaimed as a marketing advantage on the one hand, this high variability represents, on the other hand, a considerable challenge for production, processing, and final application of leather. In contrast to materials manufactured in continuous roll-to-roll production processes, such as textiles and coated textiles, the discontinuous production and processing of leather into high-quality products requires a particularly knowledge of the material. The collagen-based hierarchical 3D structure of the animal hide is essentially retained in leather manufacturing. Affecting the properties of the finished leather, the native structure causes a material-inherent large variance and pronounced anisotropy of the physical properties (Meyer2019) due to (micro-)structural variations (Haines1974; Reich2000) depending on species, race, gender, age, husbandry conditions, as well as individual body parts and tanning processes (Daniels2007). So far, it is not possible to determine the structure-related variance of physical material properties over the entire skin using the available standardised physical-mechanical test methods, as these always involve destroying the test material. Statements regarding the reasons of specific behaviour of end products that can be traced back to the structural parameters of the starting materials can only be made to a limited extent based on currently established methods. In the long term, however, suitable realistic multi-scale models for material simulation could provide a solution. These aspects increase the need for appropriate methods on leather qualification and structure-related information for specific leather applications.

Within the paradigm of industrial mass production, it becomes imperative to employ non-destructive computer-aided design to conceptualise components and to undertake numerical assessments of their physical-mechanical behaviour in advance. However, the utilisation of finite element method (FEM) and computer-aided design (CAD) software solutions to the surface material leather still remain absent commercially. The respective design and construction departments in processing industries neither have standard methods for full characterisation of leather materials nor possess suitable methods for reliably predicting the performance of leather. The definition of the necessary material qualities in the course of component design and combination is therefore still based on the separate collection and manual evaluation of a large amount of individual data from physical and microscopic examination methods.

Basic steps for modelling the leather structure had been successfully developed in previous projects (Bittrich2014; Godehardt2017; Dietrich2021). In an initial approach, the geometric properties of the leather microstructure were characterised on the basis of x-ray computed micro-tomography (µCT) data sets without automatic processing (segmentation) of the structural information being possible (Bittrich2014). These limitations were resolved in a subsequent research project (Godehardt2017). The further development of the segmentation algorithms allowed automatic segmentation and processing of the µCT data. This was established in a new procedure that makes it possible to objectively record the hierarchical leather structure on the basis of image processing methods operating automatically. As a result, large number of relevant structural parameters can be extracted from µCT images and used to characterise and compare leather structures (Dietrich2021).

In consideration of the aforementioned development statuses, the objectives of the follow-up project are twofold. Firstly, the relevant leather structure parameters are to be extracted using the previously developed novel methodology for spatially resolved structural analysis of leather based on µCT. Secondly, the macroscopic properties of leather were to be determined using multi-scale simulation applications based on a realistic model. The objective of the present study is to quantify the correlations between microstructure and properties on the basis of parameterised hierarchical structure data sets. This is to be achieved using multi-millimetre-scale simulation operations. Consequently, a methodology for the non-destructive quantification of macroscopic physical leather properties is to be developed. The aim is to provide a well-founded prediction (simulation) of the typical material behaviour of entire leather hides, with particular reference to the elongation and relaxation properties resulting from the force and tensile stresses applied. Together with experimentally determined (visco-)elastic and relaxation properties of fibre-bundles, the network model yields a meso-structural simulation tool for comparing several species, races, as well as individual body parts and tanning processes.

2 Materials and Methods

2.1 Tensile Experiments on Leathers and Fibre-Bundles

The investigations were conducted on leather samples and fibre bundles sourced from the back, flank, and axilla of metal-free, vegetable-tanned, non-dyed bovine hides of the Simmental breed (leathers L1 and L2), the Zebu breed (leather L3), and industrial syntans-tanned bovine hide (leather L4). The processing of leathers L1, L2 and L3 was executed in FILK's tanning facilities. Leather L4 was acquired commercially. The leather samples were reduced to their reticular layers by splitting off the grain and papillary layers. The conditioning and preparation of the sample followed the ISO 2418 and ISO 2419 standards.

Light and electron microscopic imaging techniques of complete leather structure areas, as well as selected substructures and individual structure elements like collagen fibres and collagen fibre-bundles, confirmed the hierarchical nature of the leather structure, as well as gradients in density (Figures 1 and 2). The weave angle, density, orientation, fibre-bundle thickness, and fibre-bundle shape can be deduced for a qualitative, rough structure description. It is evident that the intrinsic fibre-bundles appear to be wavy. The material exhibits signs of crimping. However, it should be noted that these techniques do not permit the access of actual parameterised values (Figure 3). Consequently, samples for µCT measurements are obtained from each leather L1 – L4 and all sampling positions (back, flank, axilla) for subsequent analyses.

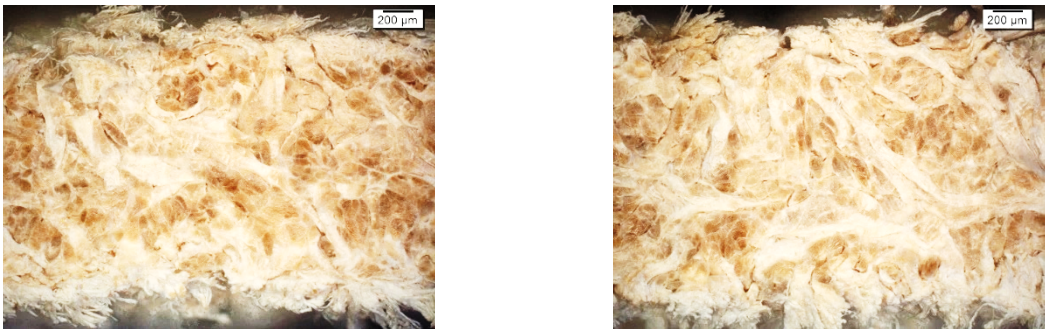

Figure 1 Light microscopic images of split leather samples from back parts: (left) Simmental breed leather L2 and (right) Zebu breed leather L3.

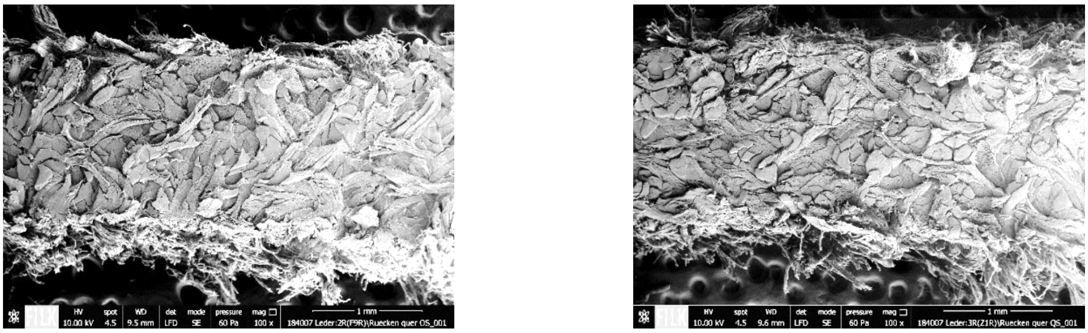

Figure 2 Electron microscopic images (secondary electron signal) of split leather samples from back parts: (left) Simmental breed leather L2 and (right) Zebu breed leather L3.

Macroscopic bulk densities and thicknesses were determined according to ISO 2420 and ISO 2589, respectively. For leathers L1 – L3, identical technological processing resulted in similar raw densities (0.51 to 0.71 g/cm^3) and thicknesses (1.35 to 1.86 mm) (see Table 1 in reference Dietrich (2021)). Results for leather L4 were in the same order of magnitude. Tensile strength σ [MPa] and elongation e [%] were measured at maximum force F_{max} [N]. Young's modulus E [MPa] are recalculated as a slope of the linear part of the stress-strain curves.

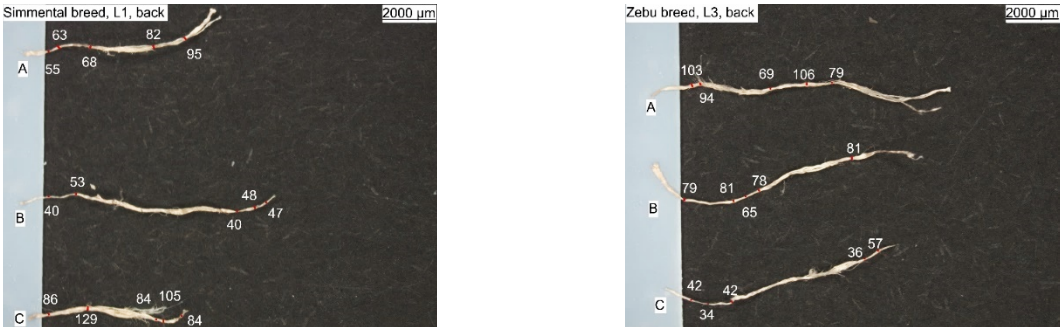

Figure 3 Light microscopic images including dimensioning of collagen fibre-bundles, extracted from (left) back of Simmental breed, L1 and (left) back of Zebu, L3).

The individual micro-mechanical stress-strain properties of collagenous fibres and fibre-bundles were measured using a test stage of Kammrath & Weiss GmbH (Figure 4), in order to parameterise their significant impact on the macroscopic behaviour of leather materials. To this end, collagen fibre-bundles were manually separated from the back, flank, and axilla of leathers L1 – L4. The fibre-bundles were imaged by light microscopy in order to ascertain their cross-sectional shapes and diameters (Figure 3). The thicknesses (or diameters, respectively) were determined as averaged values (see Table 2 in reference Dietrich (2021)) of three to five measurements taken along the length (Figure 3). It is well known that fibre-bundles vary in terms of length and fibrousness (*e.g.*, compact or loose), though not to a significant extent in diameter (see Table 2 in reference Dietrich (2021)).

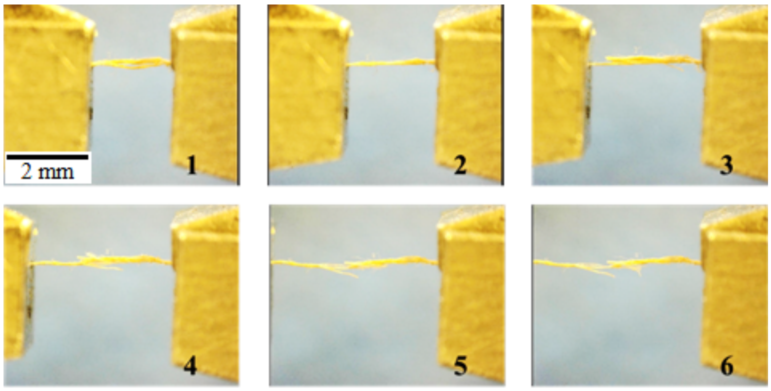

Figure 4 Illustrating uniaxial tensile test on a collagen fibre-bundle: (1) Initial state; (2) increased load causes fibre-bundle straightening and elongation; (3) slippage of fibres and beginning of fibre breakage; (4, 5) progressing sequential fibre breakage; and (6) complete breakage of fibre-bundle (Dietrich2021).

Typical force-elongation diagrams for loaded fibre-bundles are presented in Figure 5 for L1 – L4 on samples from the back and flank, respectively. The slope's characteristics depend on the sample position within the hide, but not on animal or tanning type. Consequently, elastic moduli for the fibre-bundles are similar, although the strains are higher for L4 17 – 20 % compared to 5 – 6 % for L1 – L3 (see Table 2 in reference Dietrich (2021)).

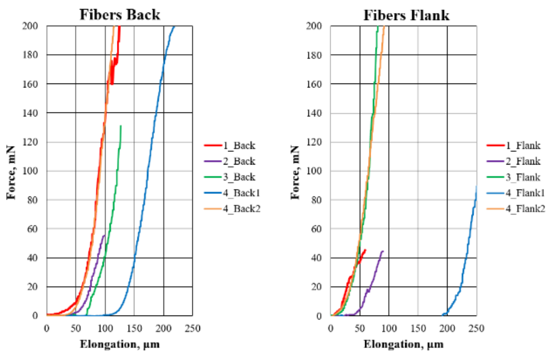

Figure 5 Comparative results of real tensile tests on fibre-bundles with elongations of 5 – 6 % (L1–L3) and up to 17 – 20 % (L4) corresponding to the strength limit: (left) back and (right) flank (Dietrich2021).

Dumbbell-shaped leather specimen from the back, flank, and axilla were tested according to ISO 3376 (Figure 6). Leathers L1 – L3 exhibited almost similar tensile strengths and elongations, while the respective values for leather L4 were slightly higher (see Table 3 in reference Dietrich (2021)). In general, the macroscopic tensile strength decreases in the order of back > flank > axilla. This observation can be linked directly to solid volume fraction. Typical stress-strain diagrams for the studied leathers correspond qualitatively to the curves for the fibre-bundles. Experimental results on leathers were similar to those for the corresponding fibre-bundles. Macroscopic elastic moduli of leathers were of the same order. Zebu samples (L3) had decreased axial stiffness compared to samples from Simmental breed (L1 and L2). The non-uniformity in elastic properties was less pronounced for the L4 samples which are however thinner than leathers L1 – L3. In some cases, large differences were observed for the leathers between the back, flank, and axilla.

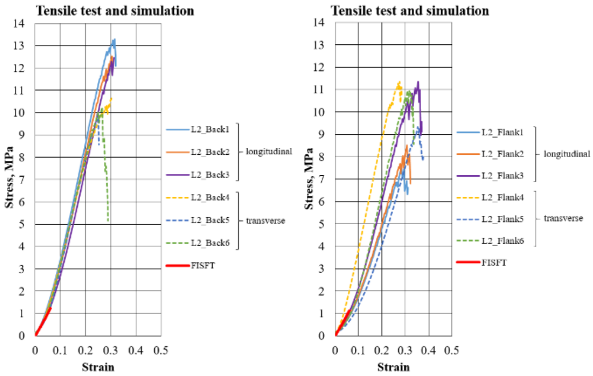

Figure 6 Comparison of experimental results with the stress simulation (FISFT) on meso-structural meshes representing leather samples of Simmental breed, L2: back (left) and flank (right). Continuous red-coloured thick lines represent results of numerical simulations; continuous thin lines are experiments for the longitudinal direction, and dotted thin lines for the transverse direction of the samples.

2.2 Micro-Computed Tomography of Fibre-Bundle Networks

Micro-computed tomography for vegetable tanned leather was successfully applied (Bittrich2014; Godehardt2017). A custom-made CT device featuring a Feinfocus FXE 225.51 x-ray tube with maximum acceleration voltage 225 kV and maximum power 20 W and a Perkin Elmer flatbed detector XRD 1621 with (2.048 x 2.048) pixels were used. Imaging parameters were optimized to capture the low absorbing meso-structure optimally. The tube voltage was low (75 kV), and integration time high (1 s). Tomographic reconstructions were obtained from 1.200 projections, each averaged over 4. Thus, the overall integration time was 4 s. The pixel edge length of 3.3 µm allowed to capture the fibre-bundles, as well as parts of their substructure.

Geometric analysis of the samples based on binary images holding the solid collagen structure as foreground and the pore space as background (Figure 7) showed the expected high variation due to the animal, breed, and position within the hide. In order to capture the local variation, four virtual sub-samples from each 3D image were cut. The back samples clearly appeared denser than those from the flank or axilla. L4 features a significantly higher surface-area per volume fraction S_V and a much lower Euler number density X_V. The latter reflects branching point density, as the Euler number is the alternating sum of connected components, tunnels, and holes in the structure. The denser the network gets, the more contacts between bundles appear per volume. This, in turn, increases the number of tunnels, and thus decreases the Euler number density.

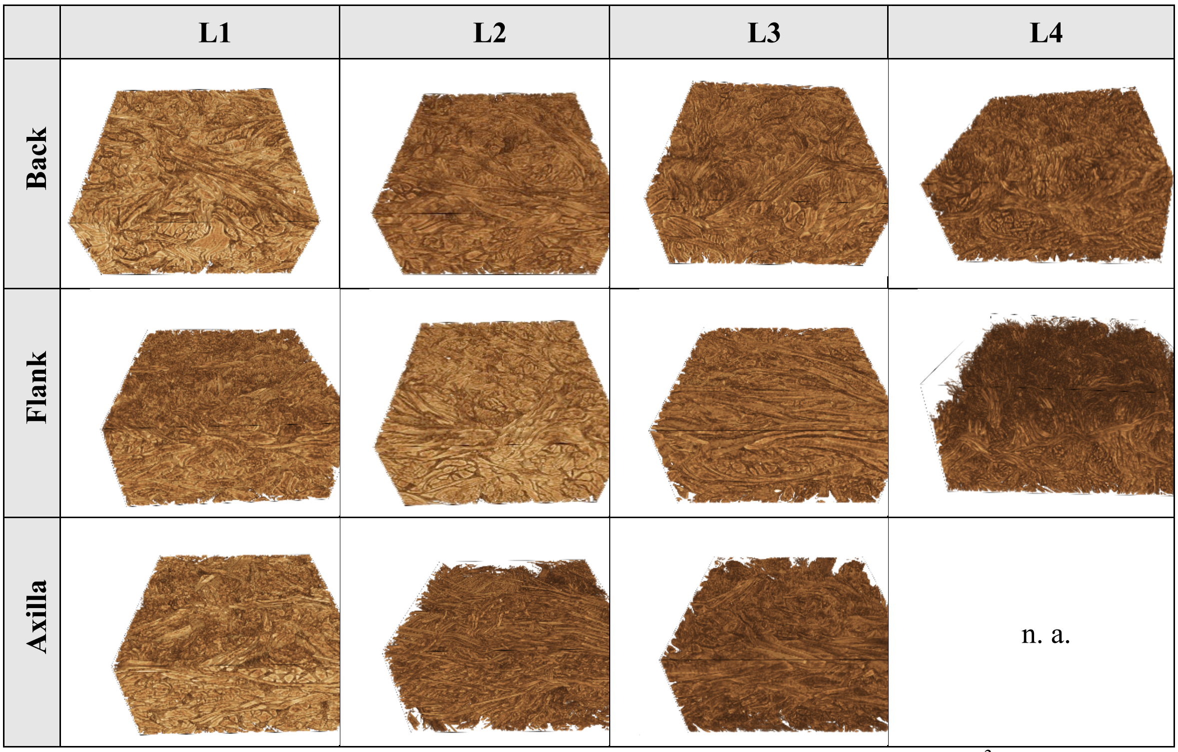

Figure 7 Renderings of (600 x 600 x 300) pixels corresponding to (2 x 2 x 1) mm^3 subvolumes of the reconstructed images of leather specimens.

2.3 Network Model

Structural modelling was required to generate geometrically simple elements including proper connectivity. The network structure of the fibre-bundles featured predominantly Y-shaped nodes. Moreover, the fibre-bundle network was forming essentially one connected component. These two observations motivated the choice of a structural model based on the edge system of a Voronoi tessellation. More precisely, the edge systems of planar Voronoi tessellations generated by macroscopically homogeneous, random Poisson point processes were rotated spatially, and interwoven. This allowed the control by a very small set of parameters, captured the microscopic spatial variation, and naturally reflected the strong prevalence of Y-shaped nodes. See Figure 8 for realizations of 2D and 3D networks, respectively. Superpositions of sufficiently many 2D networks lead to a densely interwoven spatial fibre system.

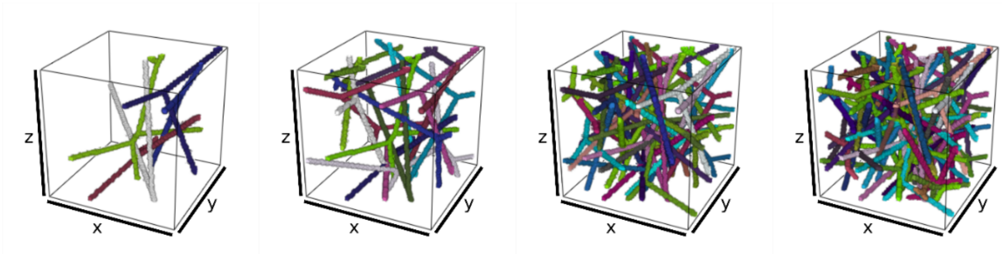

Figure 8 Rotated, superposed 2D network realizations forming a 3D network in images of 100^3 voxels. λ=10/400^2. Varying ν yields varying fibre volume rate. From left to right: ν=5, 15, 32, 45 and solid volume fractions 1, 3, 7, 10 %.

2.4 Simulation Tool

The mechanical behavior of the leathers was modelled using an in-house software tool FISFT which includes a 1D finite element method for large deformations and friction evolutions. The tool was extended to adhesion and complicated knots and wound fibres in friction contact. Moreover, it allowed to consider the influence of frictional slip evolution in local fibre-bundle knots on the macroscopic leather behavior. The meshes incorporated simple contact interactions between fibre-bundles, as well as the Y-branches found to be typical for leather. The contacts were controlled by two parameters of normal and tangential forces. Proper choice of these parameters ensured a realistic fibre reorganization of leather meshes under tensional loading in horizontal direction. In Figure 9, the relative stress distribution locally acting within each element can be seen.

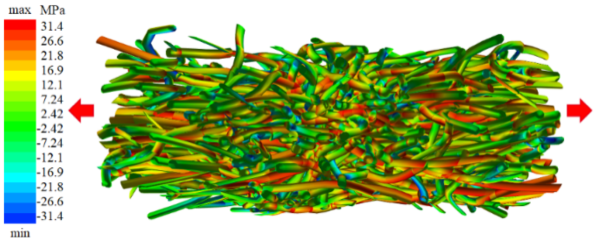

Figure 9 Reorganization of leather structure during horizontal tension test. Simulation was performed on 5 mm cubic specimen with volume fraction of 63 % (L3: Zebu back). Colours indicate axial stress values from minimal (blue) to maximal (red).

2.5 Simulation on fibres of meso-structure level

Based on the model described in Section 2.3, meshes with varying fibre volume fractions were generated iteratively. As real samples feature higher fibre volume fractions within 50 – 70 %, networks were added till the aspired fibre volume was accomplished. The stress-strain behavior of leather under tensile load was simulated on the small fibre-bundle meshes with an initial size of (5 x 5 x 5) mm^3. Experimental curves for the uniaxial tensile tests of fibres measured in 2.1 (Figure 5) were used as material input data. The results of the simulations are shown together with the experimental data for comparison for Simmental breed and Zebu, respectively. In this range, the simulation agrees very well with the experimental data (Figure 6). However, differences in scale have to be kept in mind. The leather samples can elongate much longer (about 20 – 30 %) than the fibre-bundles (5 – 10 %) without much damage just by unfolding long continuous networks of folded fibre-bundles. Extraction of samples for the μCT imaging cut the fibre-bundles, made them shorter compared to the tested fibre-bundles, and led to a reorganization of the fibres. Consequently, rather isotropic networks were obtained (loss of anisotropy).

3 Results and Discussion

Using the experimental and μCT image data as inputs for numerical simulation of leathers L1 – L3 and having verified the methodology for leather L4 of another tanning type, the main micro-structural features can be deduced by influencing the macroscopic leather behavior. The leather behavior is decisively influenced by fibre volume fraction, fibre-bundle force-strain function, and number of contacts. The latter can be changed with the tanning process. The well-known macroscopic mechanical anisotropy cannot be reproduced based on the 3 – 5 mm sample range considered here.

According to Nakahara (2020) arbitrary anisotropy can be achieved by tanning and introduction of boundary conditions (fixation and tension in certain directions). Hypothesising, on terrestrial mammals the skin is fixed at (at least) six points, four paws, and head and rear parts. These six directions determine the fibre orientation (Osaki1999) (in the pulling direction, Figure 10). Introducing new pulling directions, the anisotropy can possibly be altered. This is only possible with long contiguous and locally folded (interlaced) networks (Sharphouse1971), which unfold or disentangle themselves during deformation and reorient themselves on the geometric scale of the whole leather piece. Only by this "free-wheeling" (reorientation-unfolding) can the leather withstand much more deformation than the fibre-bundle. These hypotheses are subject of further research. Nevertheless, already now, the leather behavior on the millimetre-scale can be predicted very well, based on the fibre-bundle structure, the non-linear elastic properties of the fibre-bundles, and including the contact interaction of fibre-bundles in the numerical mechanical simulations. The simulation results agree very well with the real measurements on this scale.

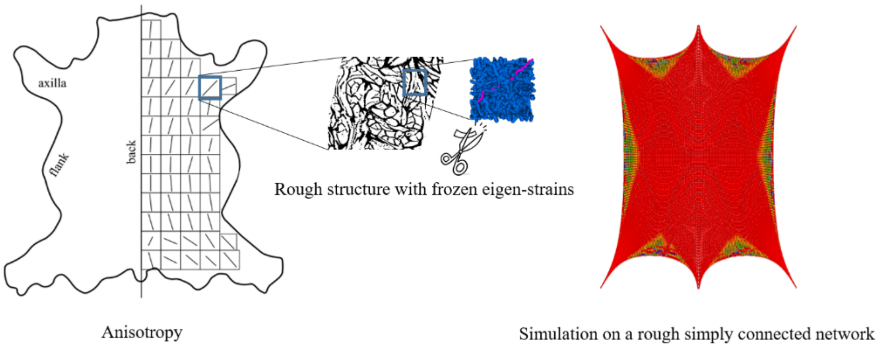

Figure 10 Hypothesis for a hide to be a connected network of locally differently prestressed fibre-bundles. (left) A schematic orientation of long fibre-bundles under introduced pulling directions; (centered) A selected long-fibre network turns to an isotropic meso-structure due to cutting during the preparation of CT-specimens; (right) Simulations of a frozen pre-stress in a fully connected network due to fixation at six directions in an animal (local fibre reorientation can be seen).

4 Conclusion

Based on high-resolution 3D geometric data from µCT images, a stochastic structure model was suggested that captures the fibre-bundle branching on the meso-scale (millimetre scale) of leather very well. Realizations of this model enable mechanical simulation, accounting for frictional sliding and visco-elastic behaviour of the leather on the basis of known (measured) behaviour of its fibre-bundles. Simulations were based on advanced mathematical modelling, reducing the dimension of contact problems and numerical techniques, including complicated mechanical issues as frictional sliding at the complex contact junctions. All simulations were validated by mechanical tests and show very good agreement on the investigated scale. The anisotropy of leather on several scales was discussed. The structural models introduced in this paper are well-suited for forecasting the leather behaviour on the millimetre scale. This paper provided and justified the hypothesis that relaxation properties of leather on this scale are defined by just the bundle structure and the relaxation time of the bundles.

Summarizing the results leads to a conjecture on the possible global network-structure of the complete leather and its local anisotropy due to frozen pre-strains in the bundles. Studying this hypothesis will help to understand the leather behaviour completely and obtain a generalized model. In the long run, a simulation tool will allow leather companies to investigate leather properties easily.

5 Acknowledgements

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