



Leather Types and Fiber-Based Leather Alternatives-An Overview on Selected Materials, Properties, Microscopy, Electron Dispersive Spectroscopy EDS and Infrared Spectroscopy

Carolina María Borlandelli; Boris Mahltig*

Hochschule Niederrhein, Faculty of Textile and Clothing Technology, Webschulstr. 31, 41065 Mönchengladbach, Germany.

***Corresponding Author(s): Boris Mahltig**

Hochschule Niederrhein, Faculty of Textile and Clothing Technology, Webschulstr. 31, 41065 Mönchengladbach, Germany.

Tel: +49-2161-186-6128;

Email: boris.mahltig@hs-niederrhein.de

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Introduction

Leather is probable one of the oldest materials which are in use of mankind even in prehistoric ages [1,2]. Nowadays leather containing products cover a broad range of different applications. Leather is used in clothes, shoes, hand bags, belts and many other things of daily use. Leather is as well used in furniture and automotive interior [3, 4]. Main reasons for this broad range of different applications are positive material properties,

especially the durability in use. Positive are also the availability and the image as a kind of premium product. Nevertheless, nowadays leather and leather containing products are part of controversial discussions. These discussions are mainly driven by two reasons. First, leather is a material from animal origin. For this, many people who are part of the vegan movement avoid the use of leather products. Vegan fashion can be also used to realize sustainable clothing [5,6]. Second, even if leather is mostly gained as by-product from meat production,



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its production has often negative impact on the environment and human health [7]. This second concern is especially valid if chromates Cr(+VI) are used during leather production [8]. While Cr(+III) has lower toxic impact, Cr (+VI) is extremely poisonous [9]. With this background, there is a certain demand for alternative materials which can replace leather in certain applications.

Conventional leather alternatives are often named artificial leather and are used as materials since decades [10,11]. These artificial leathers can be best described as composite materials or laminates. Here, a polymer coating (e.g. from polyvinylchloride PVC or polyurethane PUR) is applied on a textile fabric (e.g. a polyester woven fabric) [10-13]. It can be best described as a product from two different industries – the chemistry and the textile industry. However, especially the use of PVC materials is also under concern. PVC coatings often contain larger amounts of plasticizers which can diffuse out of the polymer matrix and can have negative impact on human health [10,14,15].

For this reason, nowadays new materials are developed and offered as possible alternative to replace leather [16,17]. These materials are also named as “trendy alternatives” [17]. In the actual report, three of these alternative materials are selected and their material composition and few properties are reported. These materials are Pinatex, Desserto and so-called mushroom leather. Pinatex can be described as composite material of plant fibers made from pineapple leaves and the biopolymer polylactide acid PLA. The fibers building up a non-woven structure and

are bound together by the PLA, which have a ratio of 20% in this material [16-19]. Desserto is also promoted as cactus vegan leather [20]. This material can be best described as laminate where a modified polyurethane coating is placed on a polyester fabric [17]. The polyurethane material is filled with at least 65% biobased particles produced from cactus plant [21]. Mushroom leather is a naturally grown material used without further coating or textile back fabric [17,22-24]. In contrast to the materials Pinatex and Desserto, for mushroom leather different suppliers offer various products fitting into this category [25,26]. The material composition of those materials is investigated by using different methods and discussed with the aim to support broad information on different alternative materials due to their composition but also to support people who are working in material analytics and have to identify new materials.

Materials and methods

Materials

For material overview altogether nine materials are selected (**Table 1**). All materials are commercially available. Four leather materials are presented (samples L1 to L4) supplied by two different companies. From each supplier a chrome tanned leather and a chrome free leather is shown. As examples for conventional artificial leather, one PVC laminate and one PU laminate are presented (samples C1 and C2). As alternative materials are chosen Pinatex (A1), Desserto (A2) and mushroom leather (A3). The material suppliers are mentioned in **Table 1**.

Table 1: Overview on discussed leather materials and alternative materials.

Sample No.	Material type	supplier	category	References
L1	Chrome Crust	Bader AG, Uruguay	Leather	[27]
L2	FOC Crust	Bader AG, Uruguay	Leather	[27]
L3	Chrome Tanned Leather	Fauk GmbH Lederhandel, Germany	Leather	[28]
L4	Vegetable Tanned Leather	Fauk GmbH Lederhandel, Germany	Leather	[28]
C1	PVC laminate	Stoffkontor, Germany	Conventional artificial leather	[29]
C2	PU laminate	Stoffkontor, Germany	Conventional artificial leather	[29]
A1	Pinatex	Ananas Anam, Spain	Alternative material	[30]
A2	Desserto	Desserto, Adriano di Marti SA, Mexico	Alternative material	[31]
A3	Mushroom leather	Grado Zero Innovation, Pangaia, Italy	Alternative material	[32]

Analytical methods

The surface topographies of investigated materials are recorded by different microscopic methods – light microscopy and scanning electron microscopy (SEM). As light microscope a VHX-600 from Keyence is used. As electron microscope a Tabletop TM3000 from Hitachi is used. The used electron microscope is equipped with an EDS unit (electron dispersive spectroscopy) Quantax 70 supplied by Bruker. The EDS spectra are recorded and by this, chemical elements on the sample surface are detected and their surface concentration is determined. Chemical elements with a surface concentration smaller 0.2 wt-% are not recorded. The chemical element hydrogen cannot be detected by EDS method. The sensitivity of the EDS method for nitrogen is small, so nitrogen is not considered for the evaluation of recorded EDS spectra. Infrared spectroscopy is done with a FT-IR spectrometer IRTracer-100 from Shimadzu.

Material characterization

Following the characteristic properties of the materials are presented and discussed. Beside material information also characteristic features are presented. These features are useful for material understanding but also for persons who are working in the field of material analytics. First, surface topographic information gained by light microscopy and Scanning Electron Microscopy (SEM) are presented. Second, EDS spectra and correlated surface composition are shown. Finally, Infrared spectra (IR spectra) are shown and main IR signals are set in relation to material composition, certain functional groups and their related vibrations.

Surface topography of materials

The microscopic images of the four investigated leather samples are presented in **Figure 1** and **Figure 2**. While **Figure 1** supports the comparison of light microscopic images in the same magnification, **Figure 2** compares SEM images recorded

in different magnifications. In fact, the typical pore structure of leather is visible which results from the animal skin [33,34]. For leather sample L3 especially brighter spots are detected on the light microscopic image and the SEM image in higher magnification. These spots could be related to applied pigments or residues from leather production process.

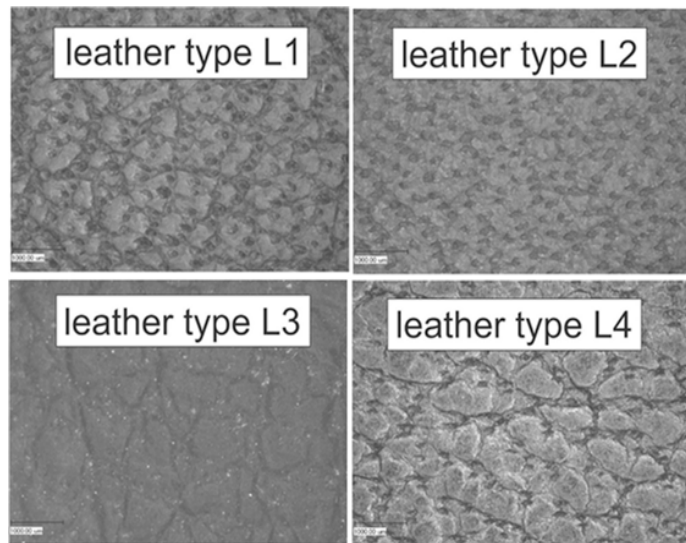


Figure 1: Microscopic images of different leather types recorded by light microscopy in reflection.

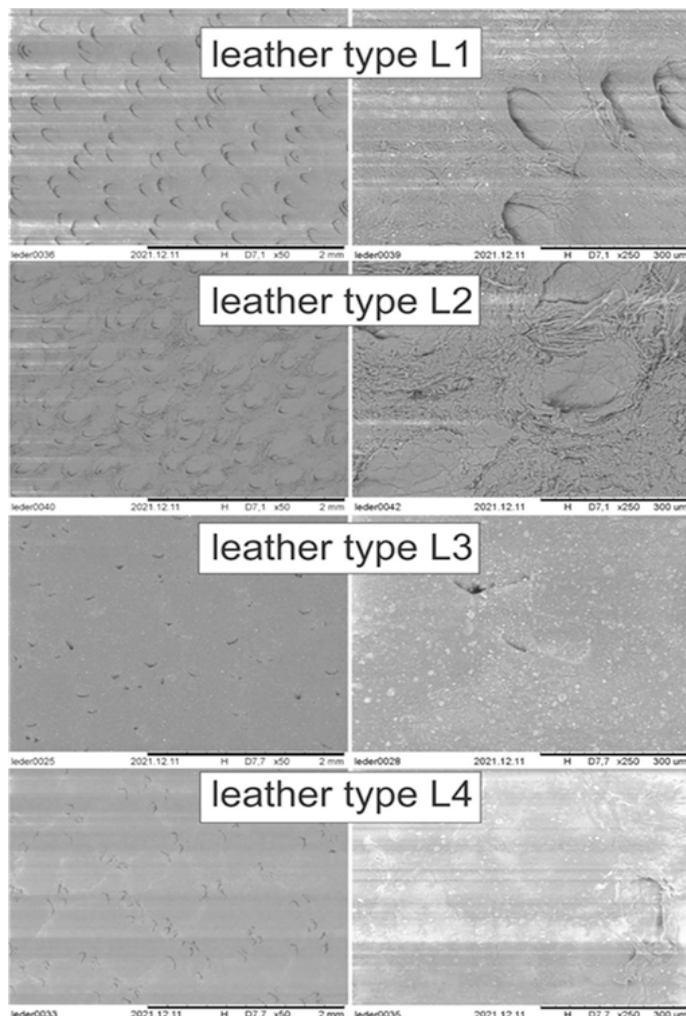


Figure 2: Microscopic images of different leather types recorded by SEM in different magnifications.

Light microscopic images are made for both artificial leather samples from top and as cross-section (Figure 3). The topography of these laminates is less structured compared to the surface of the leather samples. In cross-section the laminated polymer coating can be clearly distinguished from the textile back fabric.

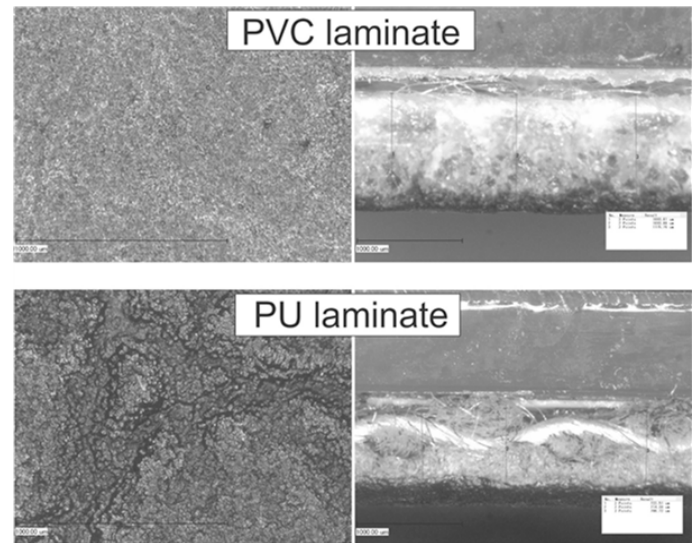


Figure 3: Microscopic images of artificial leather recorded by light microscopy – left in reflection from top; right as cross-section of the samples on a glass plate sample holder.

The SEM images of the artificial leather exhibit a flat surface topography (Figure 4). Compared to conventional leather, the recorded surface of artificial leather does not exhibit typical structural features. The SEM images in higher magnification exhibit some structural elements of low roughness.

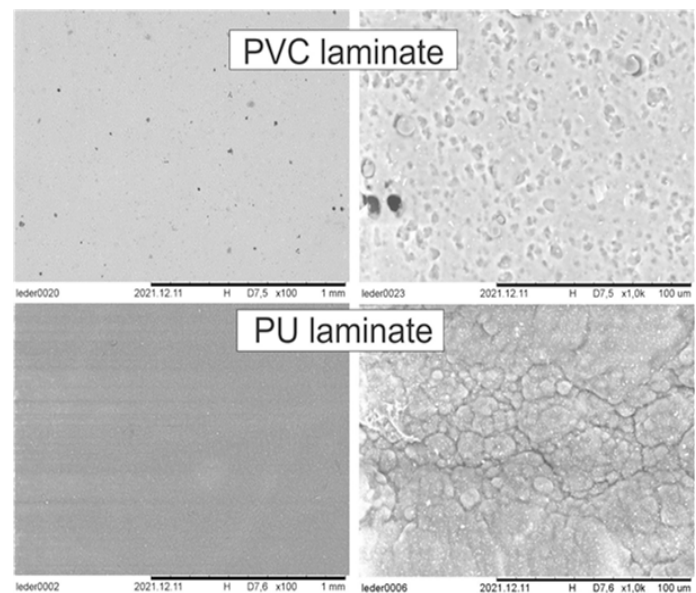


Figure 4: Microscopic images of artificial leather recorded by SEM in different magnifications.

The three alternative materials exhibit significant structural features in the light and electron microscope (Figures 5 and 6). For the Pinatex material clearly the fibers from pineapple are visible in both light microscopic images and SEM images. The polymer binder connecting these fibers is especially good recorded in the SEM image of low magnification. In contrast, the SEM image of high magnification from Pinatex offers a view on bright spots, which are probable related to filling materials added to the polymer binder. In comparison to the Pinatex topogra-

phy, the topography of the Desserto material exhibits a smoother surface (Figure 5). In cross-section, the laminate structure of Desserto is clearly visible showing the polyester fabric as backing material. In higher magnification with SEM, the Desserto surface shows also some structures of lower roughness (Figure 6). The structural similarity of Desserto to conventional artificial leather is obvious from microscopic investigations. In comparison to all actually investigated materials, the mushroom leather exhibits most characteristic and specific topographic features. Here the typical fiber structure of a naturally grown sponge is visible (Figure 6).

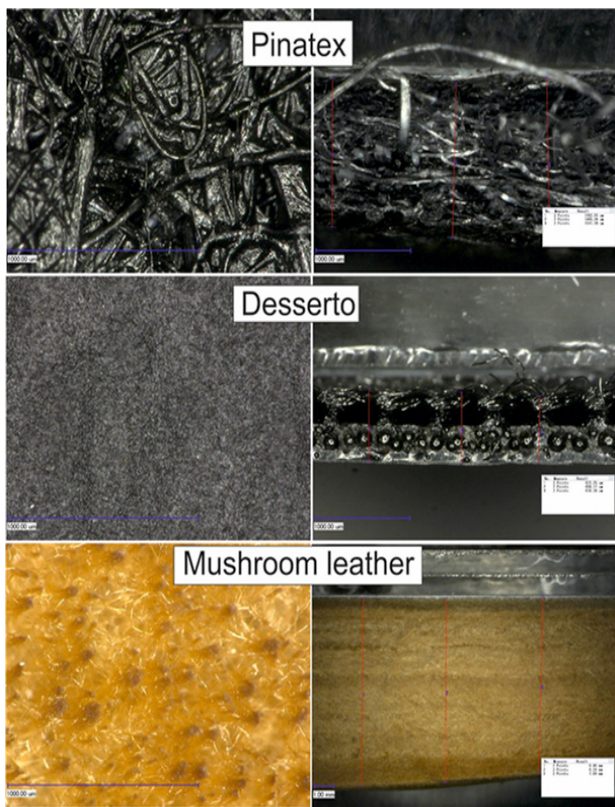


Figure 5: Microscopic images of alternative materials recorded by light microscopy – left in reflection from top; right as cross-section of the samples on a glass plate sample holder.

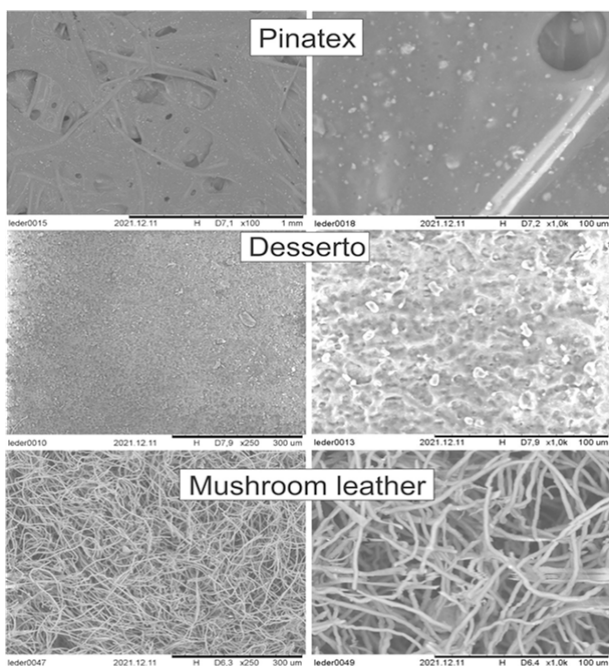


Figure 6: Microscopic images of alternative materials recorded by SEM in different magnifications.

EDS spectra and surface composition

The EDS spectra of the four investigated leather products are presented in Figure 7. The related determined surface concentrations of chemical elements are given in Table 2. By view on these data, it is clear there is a broad variation of surface composition of leather materials. This broad variation is related to type of detected chemical elements and there determined concentration. In fact, it can be stated that there is no simple typical surface composition of leather. All investigated leather samples contain carbon in a range of 52 to 70 wt-%, oxygen (range 26 to 37 wt-%), silicon (0.3 to 2.7 wt-%) and Sulphur (0.3 to 2.7 wt-%). The three elements carbon, oxygen and Sulphur are related to the animal origin of leather-built up by polysaccharide and proteins. Proteins contain as well nitrogen in relevant content. However, the EDS method has a low sensitivity to nitrogen, so it is not considered here. The determined silicon can be the result and residue of treatments during leather production processes. The leather types L2 and L4 are supplied as chrome free leather and for them no chrome is detected. For the leather types L1 and L3, chrome is determined but the content of observed chrome is strongly different with low content of only 0.2 wt-% for sample L3 and high content of 2.5 wt-% for sample L1. The low content of 0.2 wt-% chromium is near the minimum of possible detectable concentration. The leather L3 contain also titanium and iron, which can be related to applied color pigments, like iron oxide black or red [35]. Alternatively, the presence of titanium can be explained by residues from tanning with titanium (IV) salts [36].

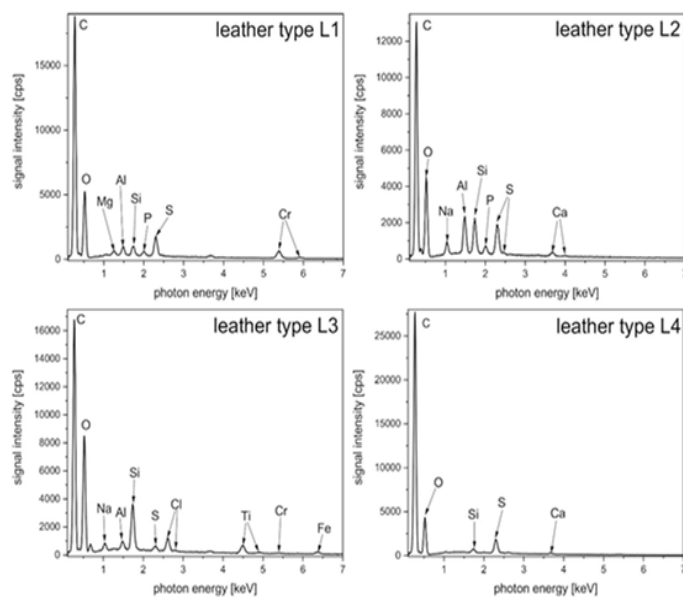


Figure 7: EDS spectra of different leather types.

The EDS spectra of both artificial leather types are presented in Figure 8. Table 3 shows the related surface composition calculated from these EDS spectra. The EDS spectrum of PVC laminate is dominated by the strong signal dedicated to chlorine (Figure 8). The sensitivity for the EDS method for the chemical element chlorine is high, so even if the chlorine concentration is lower than the carbon concentration, the EDS signal for chlorine is clearly dominant [37]. The presence of carbon and chlorine in the PVC laminate is related to the chemical structure of polyvinylchloride PVC (compare Scheme 1). Additionally, oxygen is determined in an amount of 8 wt-% and small amounts of magnesium, silicon and calcium. The presence of these chemical elements is probable related to the use of filling materials in the PVC coating. Such filling materials can be for

e.g. calcium carbonate CaCO_3 , magnesium carbonate, MgCO_3 , different silicates or particles from natural stone [38-40]. For production of antibacterial active PVC coatings on textiles also the use of calcium oxide CaO , magnesium oxide MgO and zinc oxide ZnO is reported [41]. The determined oxygen can be also related to the presence of plasticizers as additives for soft PVC. As plasticizer often, phthalate compounds are used containing oxygen as part of ester functional groups [42].

The main components of the investigated PU laminate are carbon and oxygen (**Table 3**). Carbon with a content of 65 wt-%

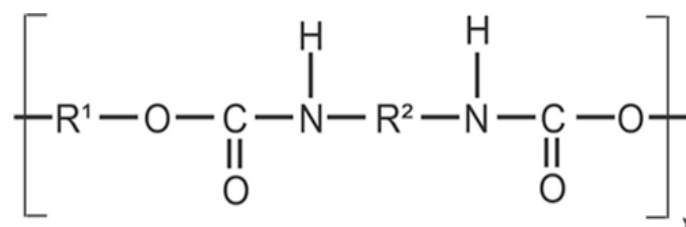
clearly dominate the EDS spectrum (**Figure 8**). Due to the polyurethane general structure (compare Scheme 2), also nitrogen should be present in the PU laminate. However, the sensitivity of the EDS method for nitrogen is low, so even in medium concentration it is not possible to identify a clear nitrogen related signal in the EDS spectrum [37]. Smaller contents of chlorine, calcium and titanium are determined as well. As source for these elements the use of calcium carbonate or titanium dioxide as filling material or as pigment is possible. The determined chlorine could be also part of organic dyestuffs added to the laminate to realize the darker coloration of this laminate.

Table 2: Surface composition of different leather types determined by EDS method. Shown are the detected chemical elements with a minimum concentration of 0.2 wt-%.

Leather / type	Content of chemical element [wt-%]												
	C	O	Na	Mg	Al	Si	P	S	Cl	Ca	Ti	Cr	Fe
L1	61.7	31.6	---	0.4	0.6	0.7	0.3	1.7	---	0.3	---	2.5	---
L2	59.4	31.7	1.1	---	2.3	2.2	0.5	2.3	---	0.5	---	---	---
L3	52.2	37.2	0.8	---	0.5	2.7	---	0.3	1.0	---	1.4	0.2	0.8
L4	70.7	26.9	---	---	---	0.3	---	1.6	---	0.2	---	---	---

Table 3: Surface composition of different artificial leather determined by EDS method. Shown are the detected chemical elements with a minimum concentration of 0.2 wt-%.

material	Content of chemical element [wt-%]						
	C	O	Mg	Si	Cl	Ca	Ti
PVC laminate	57.8	8.0	0.5	0.3	32.4	1.1	---
PU laminte	65.2	26.5	---	---	3.4	0.9	4.0



Scheme 2: General chemical structure of polyurethane PU.

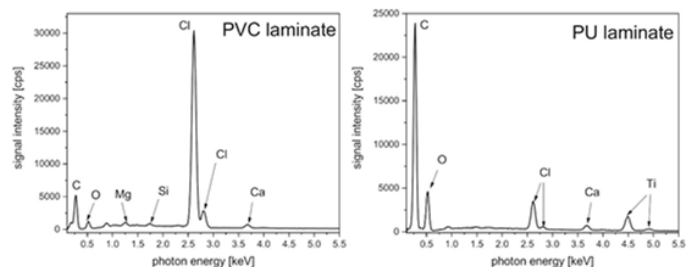
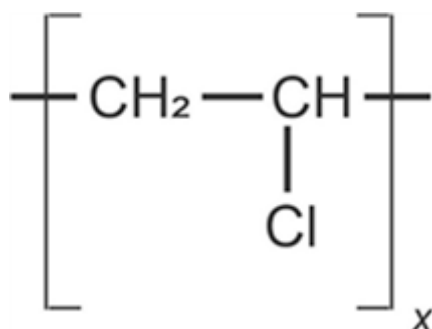


Figure 8: EDS spectra of two different types of artificial leather from PVC or PU laminate.



Scheme 1: Chemical structure of polyvinylchloride PVC.

The EDS spectra of the three alternative materials are compared in **Figure 9**. The related chemical elements are given in Table 3 together with the detected surface concentration. For the Pinatex material, the content of carbon and oxygen corresponds not only to the calculated carbon/oxygen ratio of the PLA binder. Compare also the chemical structure of PLA (Scheme 2). Each PLA repeating unit contains three carbon atom and two oxygen atoms. For this, the atomic ratio is C/O 60%/40%. Considering the atomic weight of both elements, the calculated weight ratio for pure PLA is C/O 53%/47%. The higher content of carbon determined by EDS method is probable cause by the presence of the pineapple material. Further, a small content of silicon is determined for Pinatex. This is probable related to silicate filling materials and fits to the detected pigments observed in the SEM image of higher magnification (Figure 6). From all here discussed materials, the material Desserto exhibits the biggest carbon content of 77%. Beside oxygen, no residue of other chemical elements is detected by EDS on the Desserto surface. From all investigated materials, the mushroom leather exhibits the smallest content of carbon and the biggest oxygen content. The contents of carbon and oxygen are nearly equal. This 1:1 ratio of carbon and oxygen can be explained by the polysaccharide polymers building up the mushroom filaments. Mushroom leather contains as well a small content of calcium of 1.2%. The presence of calcium can be explained by a certain mineral content of the mushroom itself or by calcium carbonate as residue from washing procedures.

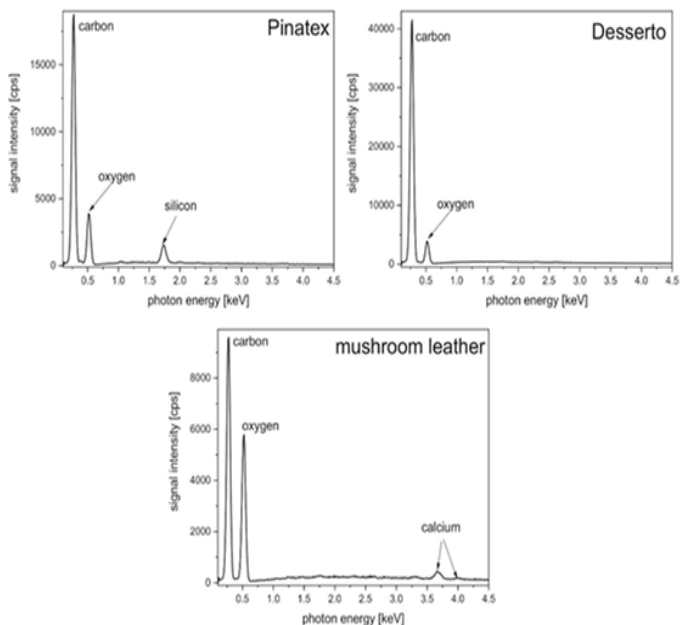
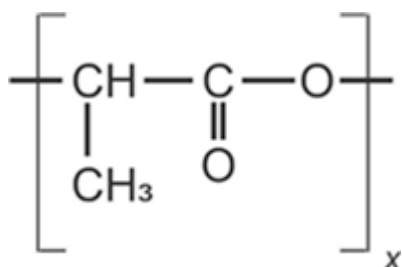


Figure 9: EDS spectra of different alternative materials.

Table 4: Surface composition of different artificial leather determined by EDS method. Shown are the detected chemical elements with a minimum concentration of 0.2 wt-%.

Material	Content of chemical element [wt-%]			
	C	O	Si	Ca
Pinatex	66.3	32.1	1.7	---
Desserto	77.0	23.0	---	---
Mushroom leather	48.2	50.7	---	1.2



Scheme 3: Chemical structure of polylactic acid PLA.

Infrared spectra

The IR spectra of both leather samples L1 and L2 (supplied from Bader AG) are compared in Figure 10. These spectra exhibit peaks at 2854 cm⁻¹ and 2925 cm⁻¹ dedicated to the presence of C-H stretch vibrations. The broad signal around 3320 cm⁻¹ is probable related to O-H stretch vibrations. The fingerprint area of both spectra below 1750 cm⁻¹ is for both leather samples nearly similar. By this spectroscopic method it is not possible to distinguish between chrome tanned and chromium free leather. This statement is in contradiction with an earlier report by Nashy et al., who observed a difference in the IR spectra of natural hides before and after chrome tanning [43]. The signal around 1650 cm⁻¹ is assigned to the amide I signal related to the C=O stretching vibration in the amide group [44,45]. The signals detected around 1550 and 1340 cm⁻¹ can be assigned to the vibrations of amide II and amide III (N-H bending vibrations) [45, 46]. These signals related to the amide vibrations results from the protein structure of leather as material from animal origin. The small signal around at 1744 cm⁻¹ are in contrast not

assigned to amide groups. Typically, this signal is assigned to a C=O stretching vibration of an ester group and could be probable the result from residues and products of the tanning process [47].

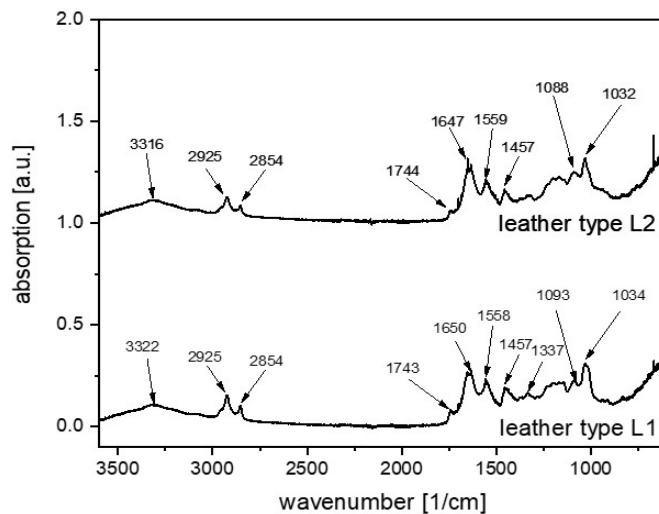


Figure 10: IR spectra of different leather types L1 and L2.

The IR spectra of leather samples L3 and L4 exhibit as well the peaks related to C-H and O-H stretch vibration. However, their signals in the fingerprint area are significantly different to the samples L1 and L2 and to each other. The main signals related to the vibrations of the amide group are as well recorded for samples L3 and L4. However, for these samples the signal around 1730 cm⁻¹ is more significant compared to the spectra of samples L1 and L2. This indicates that a different tanning process for production of leather L3 and L4 is used.

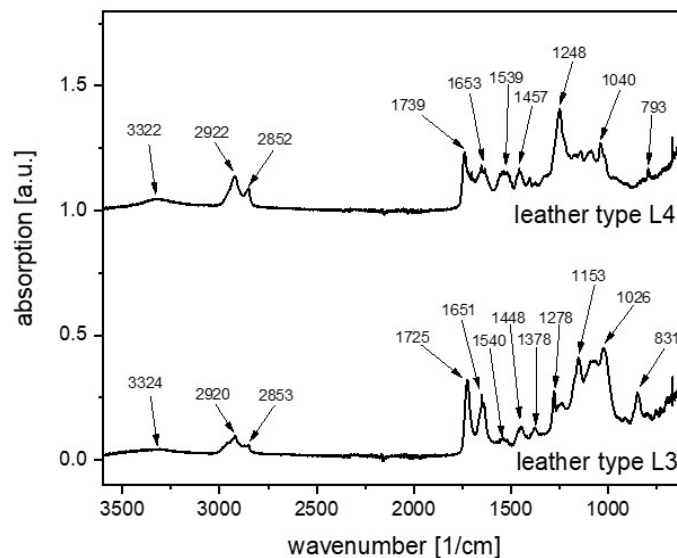


Figure 11: IR spectra of different leather types L3 and L4.

The IR spectra of both artificial leathers are presented and compared in Figure 12. Compared to all other presented IR spectra, the IR spectrum of the PVC laminate exhibit no signal in the area above 3000 cm⁻¹. PVC as hydrophobic synthetic polymer exhibits no -OH or -NH₂ groups and has no affinity to take up water from air humidity. For this, no stretch vibration for O-H or N-H can be detected. The further determined signal are not in total accordance to spectra for pure PVC polymer as reported earlier in literature [48]. Especially the peak at 1723 cm⁻¹ is not dedicated by the PVC polymer structure itself. This signal is probable related to the carbonyl C=O stretching vibration of

a present plasticizer, as e.g. Diisodecyl phthalate DIDP [49]. The peak at 1276 cm^{-1} could be assigned to a C-O stretching vibration related to the presence of the plasticizer. The medium signal at 636 cm^{-1} can be set in relation to a C-Cl stretch vibration [48, 50, 51]. The small peak at 882 cm^{-1} could be related to calcium carbonate which may be added as filling agent, because the main signal for different types of calcium carbonate are reported to appear in this area [52]. The signal around 1426 cm^{-1} could be also related to vibrations of carbonate ions [53]. However, the amount of determined calcium by EDS method is small, so the IR signals related to calcium carbonate should be as well weak and may be covered by other signals caused by vibration of the polymer PVC. The spectrum of the PU laminate exhibits a peak at 3337 cm^{-1} which is probable dedicated to a N-H stretching vibration of the polyurethane group. The signal at 2957 cm^{-1} is related to C-H stretching vibration. The three signals at 3337 cm^{-1} , 1728 cm^{-1} and 1597 cm^{-1} are related to the polyurethane group -NHCOO- (compare Scheme 2). From these signals 1728 cm^{-1} is assigned to C=O stretching vibration [54]. The both signals at 1597 cm^{-1} and 1529 cm^{-1} could be assigned to C=C stretching vibration in aromatic ring systems which building up the "hard segment" of polyurethane structure [51, 55]. The peak at 816 cm^{-1} is assigned to C-H deformation vibration of two neighboring H-atoms of an aromatic ring substituted in para position [51]. The strong peak at 1220 cm^{-1} can be assigned to the C-O stretching vibration from the polyurethane group (compare Scheme 2) [51].

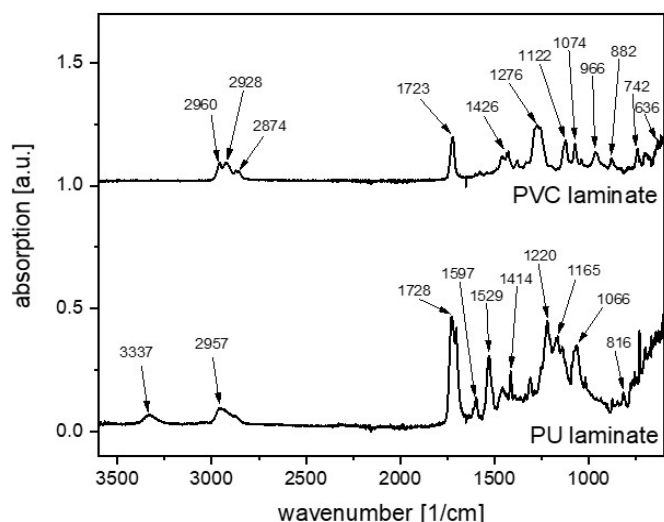


Figure 12: IR spectra of two different artificial leather types from PVC laminate and PU laminate.

The IR spectra of both alternative materials Pinatex and Desserto are compared in Figure 13. Both materials are composite materials containing at least two components a plant-based component and a man-made binder system. For this, the discussion and evaluation of these IR-spectra in relation to the chemical structure is quite complicated. The Pinatex product with a content of pineapple fiber material contains a significant amount of cellulose. Related to the presence of cellulose the signal at 3322 cm^{-1} can be assigned to O-H stretch vibrations from the hydroxy groups of cellulose. The both signals around 2900 cm^{-1} can be assigned to C-H stretch vibration from pineap-

ple material and the PLA binder. The peak at 1369 cm^{-1} and the broad shoulder of the signal at 1103 cm^{-1} are similar to signals found earlier on other pure cellulose fiber materials, and are therefore probable related to the cellulosic content in Pinatex [56]. The peak at 1717 cm^{-1} is assigned to C=O stretch vibration and the peak at 1103 cm^{-1} to C-O stretch vibration. These vibrations can be related to the ester group of PLA but also to other components which are present in the pineapple material [57]. Compared to the material Pinatex, the discussion of the spectrum from Desserto material according to the components is even more challenging. It is stated that Desserto is a composite material from polyurethane binder and plant based particles from cactus origin. However, the type of used polyurethane is not disclosed and also the components from the plant based are not reported. The broad signal at 3344 cm^{-1} can be assigned to N-H and O-H stretching vibrations from the polyurethane group or hydroxy groups from the plant based component. The signals around 2900 cm^{-1} are assigned to C-H stretching vibrations. The peak at 1740 cm^{-1} is assigned to C=O stretching vibrations from the polyurethane unit. The weaker signal at 1635 cm^{-1} can be assigned to an Amide I vibration probable related to a protein component in the plant based pigments. The strong peak at 1246 cm^{-1} can be assigned to the C-O stretching vibration from the polyurethane group [51].

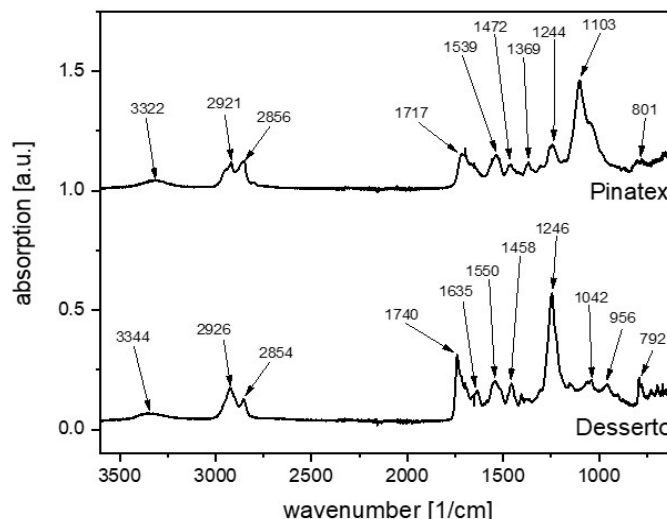


Figure 13: IR spectra of the two different alternative materials Pinatex and Desserto.

The IR-spectrum of mushroom leather is presented in figure 14. Compared to all other here presented IR-spectra, it is mainly different because it does not exhibit a signal in the area from 1700 to 1750 cm^{-1} which would be related to a C=O stretching vibration in an ester group. Mushrooms as naturally grown material are built up by a broad mixture of different molecules [58]. Main components are polysaccharides like glucans or chitin (compare chemical structures in Scheme 4) [9,58,59]. The broad signal at 3295 cm^{-1} is assigned to O-H stretching vibrations. The peaks around 2882 cm^{-1} are assigned to C-H stretching vibrations at different positions. The peak at 1636 cm^{-1} is related to Amide I band from the amide group in chitin and from protein. The strong peak at 1040 cm^{-1} is assigned to C-C stretching vibration [59].

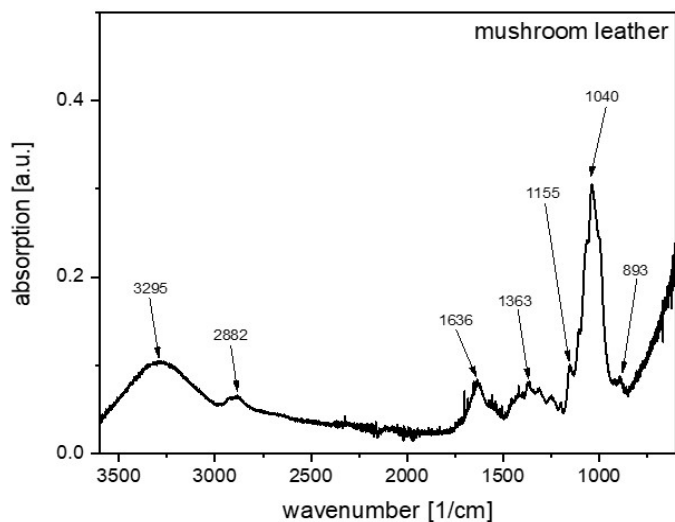
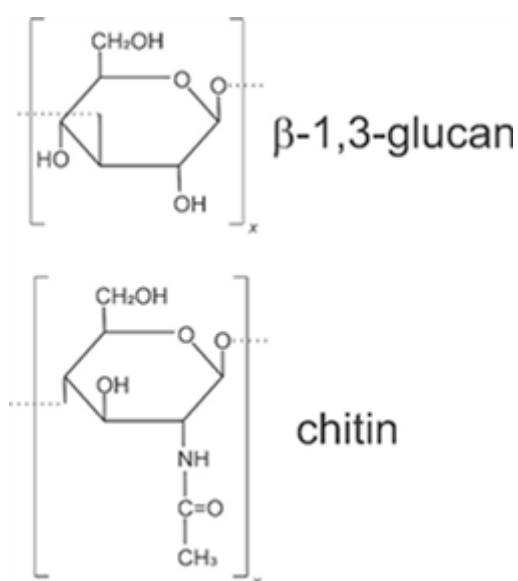


Figure 14: IR spectrum of mushroom leather.



Scheme 4: Schematic view on the chemical units of the biopolymers beta-1,3 glucan and chitin.

Conclusions

A broad range of different leather materials and alternative materials for leather are presented and discussed. The main material investigations are done by microscopy, EDS and IR-spectroscopy. A broad variety of material composition and structure is observed and is discussed in respect to supplier information and chemical background of the materials. For spectroscopic results each investigated material exhibits its unique spectral information related to the chemical composition. Beside the general review on material composition the presented data are useful for persons who are working in material analytics.

Acknowledgements

All product and company names mentioned in this article may be trademarks of their respected owners, even without labeling. No funding was supported for this article. The authors claim no financial interest or conflict to the mentioned products or companies.

Problem statement

Leather is a worldwide used material with broad application in different market segments as clothes, shoes, accessories as

hand bags, furniture and or car interior. There is a broad range of different leather materials available related to different origin and performed production procedures. Today, the production and use of leather materials is often part of a controversial discussion. This discussion is mainly driven by two issues – the origin of the leather as animal based material and the use of not eco-friendly chemicals during leather production. Here, especially the use of chromates Cr(+VI) is an issue. With this background, the aim of the current paper is an overview on different alternative leather materials and commercially available alternative materials for replacement of conventional leather. Additional to typical product properties, especially microscopic measurements, EDS spectra (electron dispersive spectroscopy) and infrared spectra are supported and discussed. By this a view on different innovative materials is supported.

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