
Shifting towards eco-responsible construction: bio-based and bio-degradable polymers as substitutes for synthetic membranes

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Abstract

Bio-based and bio-degradable polymers are a potential alternative to synthetic polymers in the application of membrane structures. In this paper gelatine-based biopolymers were chosen as a representable substitute for commonly used petroleum-based polymers in terms of mechanical properties. In the construction industry, knowledge about the application of such biopolymers is currently limited to a few experimental trials. As evidence, empirical tests were conducted under construction material standards, focusing on the analysis of the tensile strengths of untreated gelatine specimens. The results provided insights into potential application in membrane lightweight construction systems. Furthermore, challenges lied in the uniform production of the specimens to fulfill construction standards and specially, in the improvement of water resistance. Subsequently, labor conditions for manufacturing the test specimens are specified to prevent their exposure to hazardous environmental conditions that could compromise the quality of the material. Potential solutions through a series of tests by adding additives are stated in this paper. The results mainly lead to the insight that the properties are favoring bio-degradability and, thus, have a lower environmental impact than the synthetic variant.

Keywords: biopolymers, gelatine, membrane structure, tensile strength, biobased, biodegradable; water absorption

1. Introduction

Synthetic polymers are an integral part of the construction industry. Petroleum, a common primary component of synthetic polymers, poses limiting factors such as the finite availability of fossil resources or slow biodegradation speed/persistence, with potential additional pollution at the end of the plastics lifecycle and huge CO₂ emissions during manufacturing [1]. Plastics are divided into thermoplastics, elastomers and thermosets according to the cross-linking of their organic molecular chains. Strong cross-linking of the molecules ensures high strength and durability. While non-crosslinked molecules offer the property that the material is shapeable and can be melted down [2]. The transition from synthetic polymers to bio-based and biodegradable alternatives aligns with the principles of green chemistry and sustainability. Biopolymers can primarily be understood as renewable raw materials. They can be obtained from different resources such as cellulose extracted from wood, starch from corn or gelatine derived from porcine collagen [2]. Gelatine-based polymers offer a promising pathway in this transition, given their diverse range of physical, chemical, and biological properties. Gelatine is cost-effective, biocompatible, non-toxic, mechanically strong, biodegradable, and commercially viable [3]. Hence, gelatine shows high versatility and exhibits potential for further modification and exploration depending on additional material compounds. Membrane structures, understood here in the context of technical membranes, consist of fabric and an application-oriented coating. The material is used in a thin-walled, flexible configuration. The

construction experiences loading from normal forces, specifically tension, necessitating the chosen material to have high tensile strength, thus being able to withstand significant stresses. The low weight relative to surface area allows for large spans and facilitates rapid assembly and disassembly of the structure (potentially multiple), resulting in shorter construction times. The significant lightweight potential of the structures is enabled by their high specific strength. The design and application possibilities are diverse, both structurally and architecturally. Other characteristics of membranes include simple manufacturing and low material costs. Resistance to environmental influences such as radiation and moisture, including material degradation, is also a consideration. Often, in the case of transparent membranes, the material enables light transmission, promoting natural illumination or, conversely, providing shade (heat protection) [4][5].

Current research in the application of gelatine for membrane constructions is limited, with only a few experimental trials conducted thus far. However, gelatine has been increasingly explored in the textile industry. For instance, its utilization in 3D printing has been investigated [6]. Most studies on gelatine processing and application have been conducted in external fields, such as the textile industry. While these studies provide insights into the material's behavior, their focus on small-scale applications, typically in millimeters, limits their relevance to large-scale membrane structures. In membrane constructions with gelatine, there have been a few experimental attempts [7]. However, beyond the field of the built environment, membrane constructions with other biopolymers have been explored [2][4][5]. Synthetic polymers have dominated the research of tensile structures. Membranes made from biopolymers have received limited attention, with challenges such as light transmission, water resistance, and mold susceptibility. Membranes made from gelatine are emerging but lack comprehensive literature, especially for non-biomedical applications. Despite gelatine's utilization in various sectors such as medicine and packaging, valued for its biodegradability, cost-effectiveness, and capacity to generate lightweight materials, there is a paucity of literature on gelatine cellular solids. Existing studies predominantly concentrate on biomedical applications and aerogel manufacturing, often with foam specimens constrained to centimeter-scale dimensions. While certain research endeavors delve into gelatine-composite foams for non-biomedical purposes, obstacles persist due to gelatine's susceptibility to thermal effects and tendency to shrink during the drying process [8]. Consequently, the literature does not extensively document gelatine foams for industries like construction and packaging. Although patents provide some processing and property details for gelatine porous materials, their credibility can be challenging to verify.

The research aims to find a bio-based and biodegradable substitute for synthetic membrane building materials, considering the specific properties required for membrane structures. In this context, the mechanical and building physics properties are analyzed. Potential alternative applications and further research opportunities are discussed and stated.

2. Materials and Methods

2.1 Bioplastics Materials

The applied gelatine-based bioplastic (Ga_M) consists of the basic materials gelatine (powder), glycerin (liquid, gel), and water (liquid). Derived from collagen, gelatine shares a similar amino acid composition, albeit with some variations resulting from different manufacturing processes. Consequently, the molecular structure of gelatine deviates from that of native collagen. Gelatine is essentially a linear polymer, comprising a repeating sequence of almost 20 different amino acids linked together by peptide bonds [3]. However, the precise amino acid composition of gelatine is not clearly defined [9]. Collagen is composed out of three α -chains, which form a triple-helix structure. The α -chain consists of continuous repetitions of Gly-X-Y amino acid sequences where X is mostly proline and Y is mostly hydroxyproline [10]. This triple-helix is stabilized by intra and inter-chain hydrogen bonds [9]. The material is provided in powder form for the experiment. Propantriol, commonly known as glycerin, is a trihydric alcohol. The origin can be animal- or plant-based. It acts as a plasticizer, loosening the polymer chains of gelatine and reducing stiffness by weakening the intermolecular forces. For the testing, it is provided in a liquid, gel-like form. According to publications [11][2][12][13][14][15][16] on the interaction of biopolymers with additives an

improvement in water resistance was described. Gelatine has been combined with the following additives, agar agar (GaAA_M, powder), soy protein isolate (GaSPI_M, powder) [15] cellulose (GaC_M, gel), monosaccharides like sorbitol (GaS_M, powder), mannitol (GaM_M, powder) [16] and beeswax (GaBW_M, solid). These combinations significantly improved the properties of gelatine, highlighting the versatility and potential for modification of this biomaterial (Table 3). Chitosan and cellulose nanocrystals were described as having high potential [11], however, they were not tested.

2.2 Specimens Preparation process

In preparation, all objects and surfaces were disinfected with isopropyl alcohol to prevent the entry of mold. The laboratory is not completely sterile. Due to impurities in the air or temperature fluctuations, mold formation cannot be definitively excluded. All molds were protected from liquid leakage with a lid and tape. For the production of gelatine-based bioplastics (Ga_M) as a base material, 12g of gelatine, 3.6g of glycerin, and 60ml of water were added to a metal cooking pot for each dosage. The additives may also be added at this step. The mixing ratio is based on [17] and has been improved for our purposes (Table 1). The exact proportions are crucial for material uniformity. The substances were heated on a hot plate at a minimum heating level using a silicone spatula and stirred for 30 minutes. Besides, some samples were boiled for an additional 15-30 minutes. Subsequently, impurities, bubbles, and lumps were removed from the substance and poured into the appropriate mold. For the preparation of the air-enriched sample (GaF_M), the material was mixed with a stand mixer for 60 seconds at the highest speed before pouring it into the mold. For the drying process of the specimens, two techniques were used. First, the samples are placed on a drying rack with natural ventilation and a temperature of 19°C and 60% relative humidity. The drying time reached around 10 days. Alternatively, the samples were placed into a sealed drying cabin with a controlled temperature at 15°C and 60% relative humidity. The drying time was reduced to 5-6 days. The specimens were removed from the molds after drying.

Table 1: Composition ratios of the test series.

substance	unit	Ga_M	Ga_T	GaAA_M	GaBW_M	GaC_M	GaM_M	GaS_M	GaSPI_M
gelatine	[g]	12.0	24.0	12.0	12.0	12.0	12.0	12.0	12.0
glycerine	[g]	3.6	3.6	3.6	3.6	3.6	3.6	3.6	3.6
water	[ml]	60.0	60.0	60.0	60.0	60.0	60.0	60.0	60.0
agar agar	[g]			12.0					
bees wax	[g]				12.0				
cellulose	[g]					12.0			
mannitol	[g]						12.0		
sorbitol	[g]							12.0	
soy protein isolate	[g]								12.0
dose	[-]	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0

The specimens were prepared in accordance with the test series and its standardization as precised in Tabel 2.

Table 2: Test series according to their regulation.

form	testing	DIN	geometry	measurements [mm]
F1	thermal conductivity	DIN EN 12667:2001 / ISO 8302	Cuboid	150x150x120 [LxTxH]
F2	compressive strength	DIN EN 826:2013	Cylindrical	46x69 [DxH]
F3	water absorption	DIN EN ISO 62:2008	Cuboid	10x20 [LxT]
F4	tensile strength	DIN EN ISO 527-1 / -2:2019-12	Frame	180x60x1/2/3 [LxTxH]

2.3 Tensile strength testing

The specimens, based on the base formulation (Ga_M), were prepared in mold F4 for tensile strength testing. The test was conducted according to DIN EN ISO 527-1/-2 (Table 2). After production and curing, the specimens were pressed into the required shape according to the specified DIN standard. The test series consisted of specimens with thicknesses of 1mm, 2mm, 3mm, each with ten specimens. The width and thickness of each specimen were measured at 3 points. The results include the tensile strength and modulus of elasticity (Et), obtained from the measurement results, providing insights into the potential future use of the material for tension-loaded membranes. The E-modulus was determined according to the following formula:

$$Et = \frac{\sigma_2 - \sigma_1}{\varepsilon_2 - \varepsilon_1} \quad (1)$$

Measured on strain value $\varepsilon_1=0.05\%$ and $\varepsilon_2=0.25\%$.

2.4 Transparency and water solubility testing

The aim of the test series was to analyze the transparency. It was evaluated visually by placing them over a black and white striped background. Furthermore, the aim was to improve the water resistance of the test specimens. For the test series, the test specimens were made from the base material (Ga_M) with additives. Additionally, the base specimen (Ga_M) was modified. One test specimen consisted of 100% more gelatine (Ga_T), while another test specimen was boiled for 60 minutes (Ga_S). The materials were poured into mold F3. To determine the water absorption, the specimens were assessed based on DIN EN ISO 62:2008 (Table 2). The test specimens were cut into the size of 10x20mm (LxT). The thickness was determined separately depending on the additives and previous curing. The specimen's weight was measured using a precision scale. The initial weight was represented by m_1 . The specimens were placed in distilled water at room temperature of 23°C. The weight m_2 of the specimens was measured every 5-10 minutes for 60 minutes in total. The absorbed amount of water was determined according to the following formula:

$$c = \frac{m_2 - m_1}{m_1} * 100 \quad (2)$$

2.5 Mechanical properties of gelatine-based polymers

The mechanical properties of gelatine-based bioplastics depends primarily on the mixing ratio of the individual materials. Furthermore, climatic conditions such as room temperature and humidity are factors influencing the alteration of gelatine. Upon contact with water, gelatine swells, while it melts upon heating. Glycerin controls the flexibility; more glycerin corresponds to greater flexibility, while less glycerin results in brittle behavior. Another factor is the cooking process; the duration of cooking determines the amount of evaporated water. Additionally, further mechanical treatment affects air saturation and thus the strength of the material. The last factor is the framework geometry, which influences the drying time, the likelihood of mold formation due to uneven drying, and consequently, the occurrence of localized material weaknesses.

3. Results

3.1 Tensile strength testing

For the evaluation of the experiments, the key metrics are transferred onto a stress-strain diagram (Figure 1). It illustrates the behavior of the gelatin biopolymers under a specific stress by deforming and provides information about their essential mechanical properties, under tensile stress. In Figure 1, the averaged stress-strain curves of the gelatin specimens for diameters of 1mm, 2mm, and 3mm are depicted. All curves are showing a linear-elastic slope at the beginning of the curve, resulting in a brittle deformation. The gelatin biopolymers exhibited an E-modulus of 100-128 N/mm², tensile strength at break of 10.6-13.8 N/mm² (tensile strength at yield of 9 N/mm² for 3mm specimens), strain at break of 150-185% strain, maximum stress at failure of 10.6-13.8 N/mm² at a maximum force at failure of 47-74 N. Based on this, it was observed that the mechanical properties vary depending on

the material thicknesses. The E-modulus, tensile strength, and strain at break increased with increasing specimen thickness (applies to 1mm and 2mm), while the 3mm specimen exhibited significantly lower values. An apparent correlation between the E-modulus and the specimen thickness could not be inferred. However, the maximum force at failure increased with increasing specimen thickness.

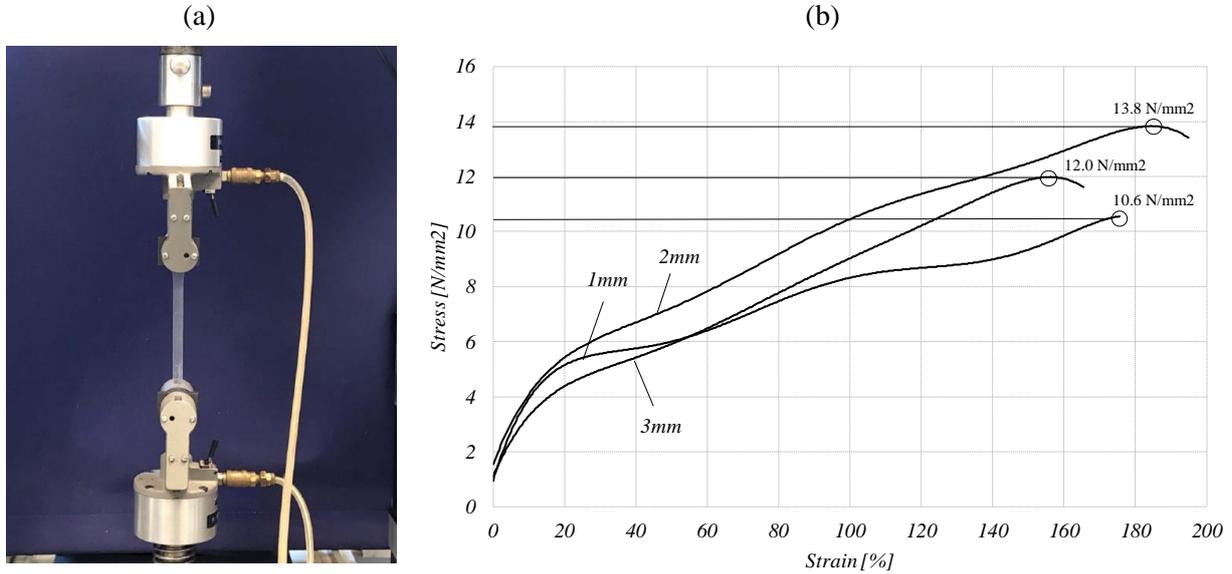


Figure 1: The results of the tensile test (a) are shown in the stress-strain diagram (b).

3.2 Transparency and water solubility testing

The appearance of gelatin biopolymers strongly depends on the additives used. The specimens consisting of the pure base recipe in various configurations (Ga_M, Ga_S, Ga_T) as well as the specimens with additive sorbitol (GaS_M) and mannitol (GaM_M) are transparent and show slight yellowish hues. Specimens with the additive beeswax (GaBW_M), cellulose (GaC_M) and agar agar (GaAA_M) were not transparent and appeared noticeably milky yellowish were as the additive soy protein isolate (GaSPI_M) appeared brownish (Table 3).

Table 3: Water absorption and transparency of the specimens.

	Ga_M	Ga_S	Ga_T	GaS_M	GaM_M	GaBW_M	GaC_M	GaAA_M	GaSPI_M
initial									
water									

The mass change of the specimens, at 60 minutes, range from 53 to 141%. The most significant water absorption with 141% was exhibited by the base sample (Ga_M) with a noticeable increase of 63% in the first 10 minutes. While the specimen Ga_T reached this point at 40 minutes and Ga_S at around 60 minutes. In comparison to specimens with additives GaS_M, GaC_M and GaAA_M who ranged after 60 minutes in an increase of 90 to 97% meanwhile GaM_M hit the 70%. The least in total incline with 53 to 54% was measured for the specimens with soy protein isolate (GaSPI_M) and beeswax (GaBW_M) with comparable results to Ga_S (Figure 2). After 60 minutes in water all specimens showing a significant change in size and thickness due to swelling while Ga_S additionally has cracked edges. The specimen with beeswax appears to have the least structural change (Table 3).

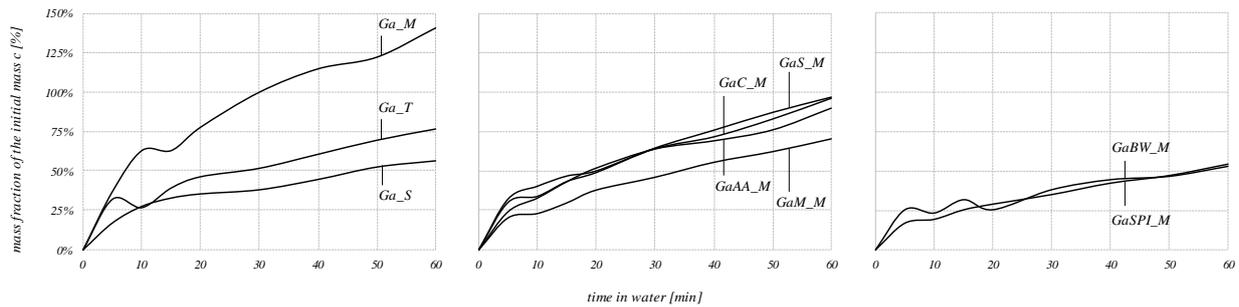


Figure 2: Comparing the results of the mass fraction of the initial mass c at a certain time in water.

The behavior of the samples upon the addition of water is attributed to its chemical properties. During the gelatine manufacturing process, collagen undergoes denaturation and loses its native structure. The collagen fibers, which normally form helices, lose their conformation during heating and partially regain it during cooling, trapping water within the mesh of chains. Consequently, the structure of gelatine differs to that of collagen, as the helices are partially reformed. This leads to different spatial arrangements and interactions among the chains. These two characteristics are influenced by the gelatine concentration, temperature, and the energy necessary for the formation of the secondary structure. Moreover, the structure of gelatine is sensitive to humidity levels, temperature variations, and the presence of various substances in the gelatine solution. Gelatine contains numerous functional groups, including -OH groups, -COOH groups, -C=O groups, and -NH₂ groups, among other, providing potential sites for a variety of molecular interactions and chemical modifications [15]. Additionally, gelatine exhibits receptivity to alterations due to cross-linkable and graftable groups [15]. As gelatine is classified as a hydrogel, its swelling behavior plays a crucial role in determining its mechanical properties [18]. Hydrogels possess the ability to absorb and retain significant amounts of water or aqueous solutions, sometimes hundreds of times their weight, without dissolving at physiological temperatures or pH levels. This characteristic is attributed to the presence of hydrophilic functional units within their network structure. Hydrogels achieve their water-absorbing capacity through physical entanglements or chemical crosslinker, rendering them insoluble in the surrounding solution. Hydrogels can gradually release approximately 95% of the water they absorbed when exposed to dry conditions and being able to rehydrate once they come into contact with water again [19]. The dissolution of gelatine in water typically occurs in two distinct steps: first, the gelatine undergoes swelling, followed by its eventual dissolution as it reaches its melting temperature. These swelling and melting steps are influenced by various external factors, such as pH, ionic strength and water temperature, underscoring the complex interplay between gelatine and its environment [9].

3.3 Additional attempts: compressive strength and thermal conductivity testing

Due to increased mixing, the material was brought into a foam-like state, prompting the possibility of further experimentation. The material was poured into different molds. The first experiment aimed to determine the state of the material in a flat form (F4). For the second experiment, the material was poured into mold F2 to test its compressive strength according to DIN EN 826:2013 using the Zwick

Zmart.Pro 10KN testing machine. The results were expected to indicate the material's potential for structural applications. There was a theoretical consideration to test the materials thermal conductivity according to ISO 8302 or DIN EN 12667 using the 'lambda-Meter EPSOOe' thermal conductivity meter following EN 1946-2 by Lambda-Messtechnik GmbH Dresden.

The first experiment yielded specimens (F4) with a top surface composed of a hardened, foam-like layer approximately 2mm thick, connected to a bottom surface made of a clear layer approximately 1mm thick (Figure 3). The production of specimen F2 could not be carried out uniformly. After 48 hours the mold was removed. The specimen consisted of an uneven top surface with a hard, foam-like layer 60mm thick, connected to a bottom surface made of a transparent layer 3mm thick with an approximate shrinkage of 10mm. Although the test specimen is completely cured, the uneven condition was a reason not to produce test specimens F1 for the DIN standard.

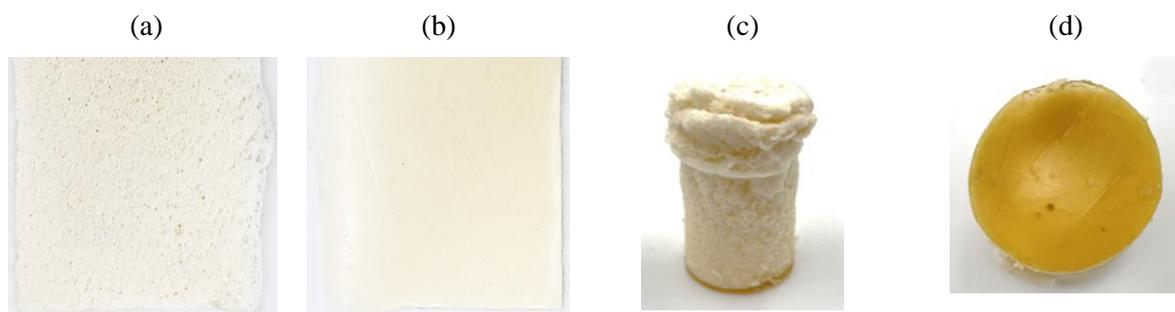


Figure 3: Visualization of the test specimens: sample (F4), top side cured foam-like (a), bottom side solid (b); sample (F2) top side cured foam-like (c), bottom side solid (d).

4. Discussion

Comparing the properties of gelatin biopolymers with synthetic polymers (Table 4), it is observed that both materials typically exhibit a linear elastic region in the stress-strain diagram. The E-modulus of the samples can be compared to that of polyethylene (PE). The variance depends on the molecular structure, crosslinking density, and chain flexibility. Both synthetic polymers and gelatin biopolymers exhibit a yield point (no yield region), followed by brittle deformation. However, the yield strength is significantly lower than that of the synthetic counterpart. The tensile strength of the tested samples falls within the ranges observed for PVC and PE.. To optimize the mechanical properties in the context of load-bearing structures, a composite material with nanomaterials (e.g. nanocellulose) could be utilized [11]. Reasons for the deviation of the results despite identical specimen formulation lie in the imperfect composition of the mixing ratios and thus possible material variations. Furthermore, unequal drying of the specimens or residues such as bubbles can impair their strength. However, environmental impacts during the tests, such as temperature and humidity, can lead to variations in the stress-strain diagram results. Understanding and controlling the behavior of the samples regarding hydration and swelling behavior is crucial, as it significantly influences the stability of the membranes under load. While it has been shown that adding additives reduces the process of water absorption, a change in state could not be avoided. Thus some additives such as beeswax could keep the initial structure nearly unchanged. This emphasizes the importance of considering a realistic application scenario while characterizing the material, such as membrane construction (Figure 4).

Table 4: Properties of synthetic polymers compared to gelatine biopolymer [2].

Polymers	density [g/cm ³]	using at temperature shortly [°C]	characteristics/ transparency/ breaking behaviour	tensile strength [N/mm ²]	E-modules [N/mm ²]
Thermoplastics					
Polyvinyl Chloride (PVC)	1.46	75-90	flexible/ transparent/ ductile	10-25	
Polyethylene (PE)	0.92	80-90	waxy/ milky/ ductile	8-23	200-500
Polypropylene (PP)	0.90	140	milky/ ductile	21-37	1100-1300
Polyethylene Terephthalate (PET)	1.37	200	transparent	47	3100
Ethylen-Tetrafluorethylen (ETFE)	1.75	220	transparent as film	35-54	1100
Polycarbonate (PC)	1.20	160	tough/ transparent/ ductile	56-67	2100-2400
Elastomers					
natural rubber		90.00	opaque/ ductile	30	
Duroplastics					
Polyurethane (PUR)	1.05	70-100	opaque/ ductile	70-80	4000
Bio-based + bio-degradable					
Polyhydroxybutyrat (PHB)	1.20			32	
Polylactic Acid (PLA)	1.21		shiny/transparent		
Gelatine (Ga)	1.60	50	flexible/ transparent/ brittle	10.6-13.8	100-128

Based on the results regarding the mechanical and building physics properties, the application of gelatin biopolymers can be discussed for areas or industries where sustainability and environmental impacts play a crucial role. The dependence on finite and non-renewable resources is reduced, resulting in a lower CO₂ footprint, primarily due to more environmentally friendly manufacturing. The biodegradability can be beneficially utilized at the end of life, potentially leading to a reduction in environmental pollution and waste generation. Moreover, the properties can be tailored to the application and its requirements through modification of the molecular structure or the use of the material as a composite. Considering the ecosystem and thus the interactions between humans and the environment/soil, biocompatibility is advantageous, avoiding undesired chemical reactions, would may be also harmful. Gelatin biopolymers are cost-effective and available on a large scale. However, the production of gelatin should also be critically examined. Plant-based biopolymers such as pectin, might represent an alternative, pending the availability of test results. Overall, gelatin biopolymers present a material that can reduce global challenges concerning resources, water scarcity, environmental pollution, and health.

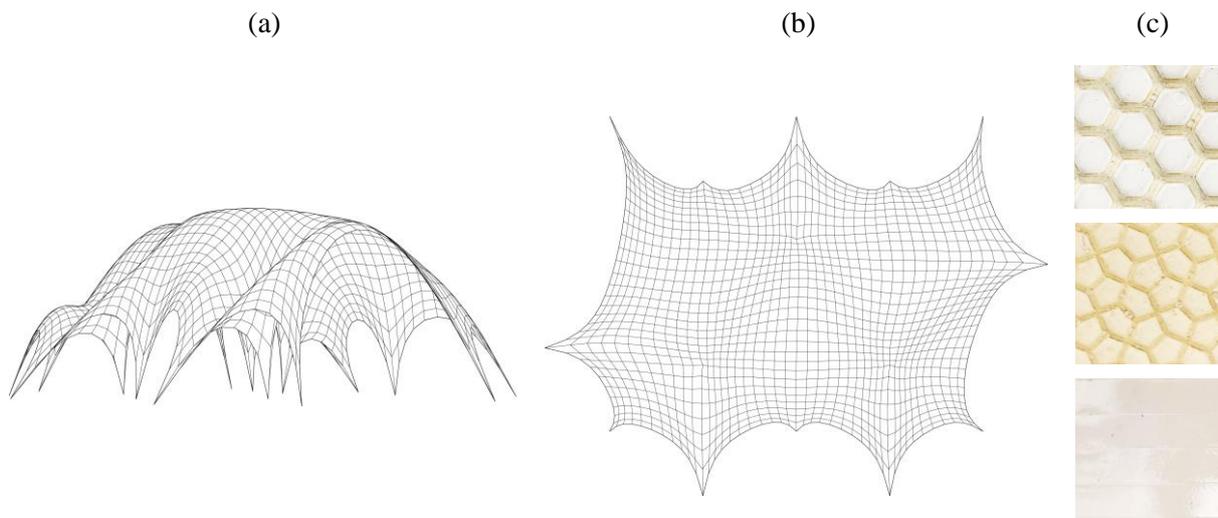


Figure 4: Possible application as membrane constructions: (a) side view, (b) top view, and (c) possible membrane structure.

5. Conclusion

The results of the test series prove the potential of bio-based and biodegradable polymers as a substitute for synthetic membrane building materials. The gelatine-based material (Ga_M) with a thickness of 2mm achieved an E-modulus of 128 N/mm² and a tensile strength at break of 13.8 N/mm² and showed the highest values among the tested test specimens. Properties such as transparency and color variation (yellow/brownish tint) can be controlled by the addition of additives. Water resistance poses a significant challenge. In the primary state (Ga_M), a mass change of 141% was observed after a test duration of 60 minutes due to the absorption of water. If the production process of the material is modified by a longer cooking time (GA_S), a change of 57% occurs. With the addition of additives such as beeswax (GaBW_M), the best results were achieved with 53%, followed by soy protein isolate (GaSPI_M) with 54%.

The transition to an eco-friendly construction by using bio-based and biodegradable polymers shows excellent potential for sustainable architecture. Gelatine-based materials are facing significant challenges. These challenges include concerns about their durability, especially their tendency to degrade over time due to environmental factors like moisture. Additionally, ensuring structural stability and strength, particularly in load-bearing scenarios, is difficult with gelatine materials. Furthermore, ethical and environmental considerations arise from the animal sourcing of gelatine, which may conflict with sustainability principles. Also, the inconsistency in properties and lack of standardization make it challenging to predict the behavior of gelatine-based materials across architectural projects. Moreover, it's essential to acknowledge that, despite the difficulties associated with gelatine-based materials, this research is a starting point for exploring other biopolymers. By tackling issues like durability and structural stability with gelatine, this study sets the stage for investigating alternative bio-based materials. This broader exploration could uncover new solutions with improved sustainability and performance for architectural use.

Acknowledgements

All presented material tests were conducted at the Institute of Building Materials Research (ibac), RWTH Aachen. Some of the tests were produced as a part of the Honors College Summer School “Structural materials for the post-oil era”.

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