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Upcycling Salmon Skin Waste: Sustainable Bio-Sequins and Guanine Crystals for Eco-Friendly Textile Accessories

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Abstract: The significant environmental impact from fashion and textile industries has spurred interest in sustainable alternatives, especially for accessories like sequins and beads, whose usage has surged post-pandemic. This study explores the potential of utilizing salmon industry waste from Chile to produce bio-sequins (BS) and guanine crystals (GC) from salmon skin. The production of BS offers a strategy to reduce reliance on non-renewable resources and support sustainable waste management, as these materials decompose naturally without harmful residues. Physicochemical and mechanical characterization of the BS by using scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), thermogravimetric analysis (TGA), X-ray powder diffraction (XRPD), and Fourier transform infrared spectroscopy (FT-IR), evaluated their feasibility for textile, design, and fashion applications. Additionally, GC were extracted from salmon scales using less hazardous solvents such as acetone, ethanol, and acetic acid, and subsequently immobilized on the BS for decorative purposes. Notably, tensile mechanical properties of the BS improved up to 75% after guanine decoration and exposure to simulated environmental factors like UV radiation. This work addresses the dual challenge of pollution and resource depletion, demonstrating that BS from salmon skin offer an eco-friendly alternative. It underscores the importance of adopting sustainable practices throughout the fashion industry's production chain.

Keywords: biobased materials; bio-sequins; design; guanine crystals; salmon skin; waste valorization; upcycling; sustainability

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1. Introduction

The depletion of natural resources and the alarming rate of environmental pollution have compelled the fashion and textile industry to undergo significant changes toward sustainability [1–3]. Historically, fashion has been recognized as one of the most polluting industries, only seconded by the energy sector [4,5]. The extensive use of synthetic fibers, chemicals, and water, coupled with the generation of textile waste, makes it a considerable environmental burdening need for sustainable practices. This reflects broader societal changes in lifestyle, impacting choices related to what we wear, buy, and discard. At the same time, the global salmon industry has seen significant growth over the past few decades [6]. In Chile, which is the world's second-largest producer of farmed salmon, this industry plays a vital role in the economy, with exports ranking as the third-largest sector of the country's productive output [7–9]. However, this rapidly resulted in considerable environmental challenges. Salmon farming produces an estimated 400,000 tons of organic waste annually, including a large volume of fish skin and scales [10]. This waste poses environmental management challenges. Nevertheless, the valorization of these biological by-products presents a significant opportunity: transforming unwanted waste into valuable, biodegradable materials could contribute to more sustainable practices in both the aquaculture and fashion industries.

Sequins, small decorative accessories often found in garments and textiles, are typically manufactured using non-biodegradable plastics such as polyvinyl chloride (PVC) and polyester. Although sequins add aesthetic value to clothing, they also contribute significantly to the growing crisis of microplastic pollution; it is estimated that over 170 million tons of microplastics are currently floating in the world's oceans [11]. These plastic particles, smaller than 5 mm in diameter, are detrimental to marine ecosystems. They are often ingested by marine organisms, ultimately entering the food chain and affecting both wildlife and human health through bioaccumulation and biomagnification [12–14]. The manufacturing, use, and disposal of plastic sequins eventually release microplastics to the environment which is a considerable challenge for waste management systems due to the size of these particles, that makes them difficult to filter or remove. Furthermore, the synthetic materials used to make sequins are derived from fossil fuels, leading to the depletion of non-renewable resources and the emission of greenhouse gases during production. In light of these environmental concerns, reducing or eliminating the use of polluting materials in textile accessories is critical.

One promising solution lies in the development of biobased plastics, which are derived from renewable resources and designed to be biodegradable. These materials aim to reduce the environmental footprint of synthetic plastics by decomposing into harmless substances under natural conditions. In recent years, advancements in biopolymer science have led to the creation of various synthetic biodegradable plastics, such as polylactic acid (PLA), polyhydroxyalkanoates (PHAs), and starch-based polymers, which are already being applied in the packaging and textile industries [15–19]. However, these plastics present a great challenge for tunning their mechanical properties with scalability to be effectible biodegraded [20]. For instance, while materials like PLA have found widespread use, they may still require industrial composting facilities to decompose efficiently [21,22]. Moreover, synthetic biodegradable plastics often exhibit lower tensile strength and flexibility as compared to their petroleum-based counterparts, limiting their application in certain industries like fashion and textiles. Despite these limitations, research focused on biobased materials continues prioritizing sustainability [23–25]. In the case of fashion and textiles, the valorization of waste to create biobased materials is a promising approach to reduce environmental impact. For example, plant-based sequins made from natural fibers have emerged as eco-friendly alternatives to synthetic plastics in textile applications [26,27]. Research has shown that materials made from cellulose, starch, or other natural fibers, feature a smooth surface and can be processed into textiles or decorative elements. Moreover, fish skin, scales, and bones, typically discarded as waste in the salmon industry, can also be repurposed into biobased textiles and accessories such as sequins. Salmon skin is gaining attention as a valuable by-product in creating biobased materials due to its high collagen content, which can be used to create biodegradable films, leather-like materials, or functional bioplastics [28,29]. This not only reduces reliance on non-renewable resources but also contributes to the circular economy by transforming waste into valuable products [30]. Another aspect of this research involves the extraction of crystals from salmon scales, a practice that mimics the natural formation of these iridescent crystals in marine organisms. Guanine crystals (GC) are responsible for the shimmering, reflective properties often found in fish skin, and they offer an eco-friendly alternative to synthetic dyes or pigments commonly used in textile decoration [31–33]. Traditional methods for guanine extraction use harsh chemicals like HCl or NH_3 to isolate the crystals. While these methods can be effective, they present significant environmental and safety concerns. For instance, HCl is corrosive and can lead to harmful emissions if not properly handled. Similarly, NH_3 is a volatile compound that can be hazardous to both human health and the environment if released during the extraction process. To address these challenges, greener extraction techniques have been explored by using more ecofriendly solvents [34]. These methods are not only less harmful but can also be reused in a closed-loop system, further minimizing waste and environmental impact. Moreover, the extracted crystals could be immobilized onto biobased sequins, enhancing their aesthetic appeal while maintaining eco-friendly properties.

As summarized in Figure 1, this research aims to develop biodegradable sequins and guanine crystals from waste generated by the salmon industry, particularly fish skin and scales. The goal is to provide an eco-friendly alternative to plastic sequins that contributes to reducing pollution while promoting responsible waste management in the fashion industry. The physicochemical and mechanical properties of the biobased sequins will be thoroughly characterized to assess their suitability for use in textiles, and their potential to decompose naturally in the environment will be evaluated. Additionally, this study will explore the use of less hazardous solvents in the extraction of guanine crystals, providing a more sustainable approach to textile decoration. By combining the valorization of organic waste with biobased material production, this research contributes to the ongoing efforts to reduce the environmental impact of the fashion industry. It also highlights the importance of adopting greener processes, such as safer solvent use, to ensure that sustainable alternatives can be both effective and environmentally responsible.

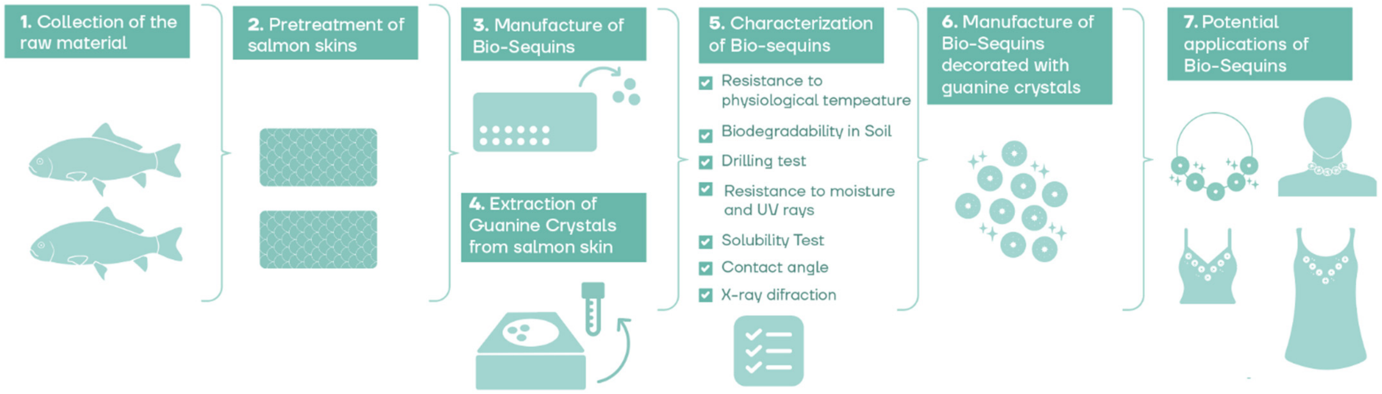

Figure 1. Development and characterization of a biobased sequin decorated with guanine crystals from waste generated by the salmon industry.

2. Results and Discussion

2.1. *Characterization of the Bio-Sequins*

2.1.1. Resistance to Physiological Temperature

Physiological temperature tests are relevant, as the bio-sequin is expected to withstand the human body temperature, which ranges between 37 and 42 °C. This test ensures that the quality and efficiency of the biobased material will not deform, crack, or brittle upon exposure. As such, these results will assess the quality and comfort standards that biobased materials should meet. Moreover, this characterization is essential to determine the durability of this garment upon human skin contact, confirming the resilience and suitability of the biobased material for clothing and textile applications. Figure 2 illustrates the bio-sequin before and after exposure to the aforementioned temperatures.

From Figure 2, there can be seen that the biobased material did not undergo significant changes in its structure upon exposure to 37 °C and 42 °C for 12 and 24 h. These findings suggest that the developed bio-sequin is capable of withstanding physiological temperature for extended periods. Since body temperature typically falls within 36–37 °C and can rise under certain conditions, the test within the 37–42 °C range ensures that the material remains stable under realistic use situations, such as when in contact with skin or under warm environmental conditions. Furthermore, these results provide evidence of the quality and resistance of the material, meaning that it is unlikely to degrade or deform during normal wear, making it suitable for potential applications in clothing and textiles. This aligns well with the response of other biobased materials exposed to body temperature [35].

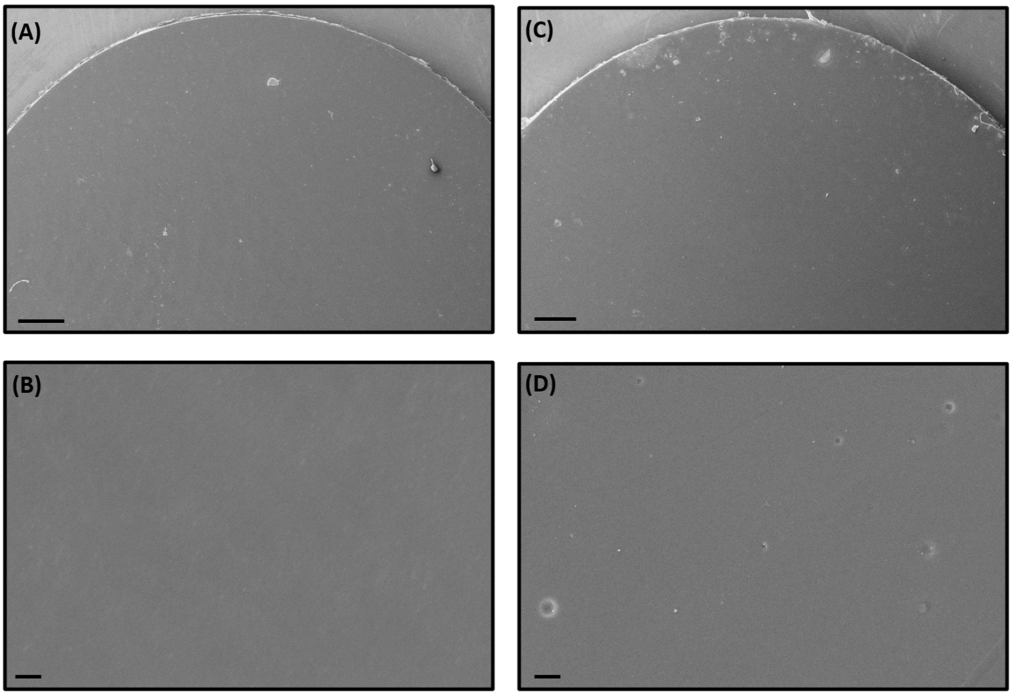

Figure 2. SEM micrographs of the BS before (**A**,**B**) and after (**C**,**D**) exposure to physiological temperature (37–42 °C). Scale bar of 1 mm (**A**,**C**) and 2 μm (**B**,**D**).

2.1.2. Biodegradability in Soil

Biodegradability tests are of vital importance, to ensure that the biobased material complies with the sustainability prospect intended in this research. This assay provides information concerning the decomposition of the bio-sequin in soils at different intervals. Biodegradation is also assessed to ensure that the biobased material is environmentally friendly, preventing the accumulation of non-biodegradable waste, promoting a responsible life cycle and envisioning the possibility of soil nourishment. Figure 3 illustrates the bio-sequin after a degradation period of three months.

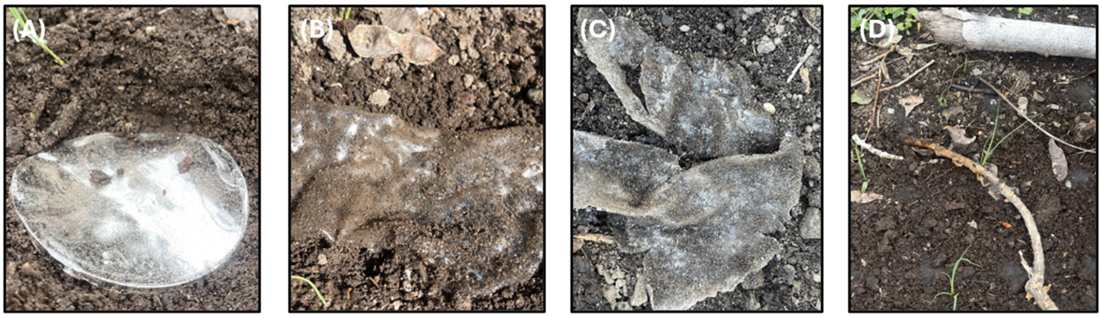

Figure 3. Mass of the developed BS after composting during a week (**A**), one month (**B**), two months (**C**), and three months (**D**) in soil.

As observed in Figure 3, biodegradability tests reveal that the bio-sequin is highly susceptible to decomposition in soils. The biobased material was buried for a period of three months, during which weight loss of the material was recorded daily. Throughout the test, it was evident that the bio-sequin underwent significant changes in its appearance. On the second day, the material became moist, soft, and acquired a gelatinous texture. These changes suggest that the material began to quickly decompose, affecting the integrity of its structure. Finally, by the third month, the bio-sequin had disintegrated completely. This indicates a high rate of biodegradation under soil conditions. As such, the environmental impact and circular life cycle of the bio-sequin can be inferred. Although biobased materials decompose at a higher rate that their petroleum-based counterparts, their effects on soil biota and seed germination must be studied [36,37].

2.1.3. Drilling Performance

Drilling performance tests were conducted to ensure that the biobased material will not brittle or crack when subjected to various drilling methods. Such methods include die-cutting or extrusion. The functionality and potential applicability of the bio-sequin depends on its ability to withstand both external (determines the shape of the bio-sequin) and internal (allows the bio-sequin to be threaded) drilling. As such, drilling performance tests can confirm the quality of the product and its foreseeable lifespan. The performed drilling performance tests are summarized on Figure 4.

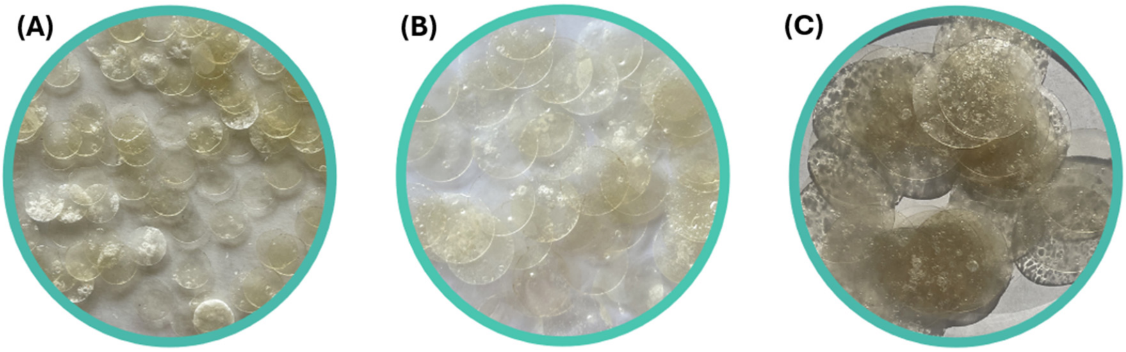

Figure 4. Drilling tests of the BS, with three variable diameters: 0.8 mm (**A**), 1.5 mm (**B**), and 2 mm (**C**).

The results indicate that the bio-sequin can be drilled without compromising its structural integrity, revealing its ability to withstand moderate tension. Additionally, this biobased material can be handled without showing signs of deterioration. Further, the bio-sequins were subjected to drilling under three different diameters. This confirmed the feasibility to develop bio-sequins of different sizes, including maxi and mini sequins, which are prominent in the market. It is worth noting that mini sequins maintained their original shape despite being harder to drill internally. Overall, the obtained results suggest that the biobased material is suitable for clothing applications.

2.1.4. Resistance to Moisture Under Storage Conditions

Moisture resistance tests were conducted to assess the ability of bio-sequins to withstand darkness, humidity, and prolonged storage periods without losing its properties. Resistance to moisture is also relevant to prevent mold formation, water penetration, and deterioration of the biobased material. The resistance of the bio-sequin to moisture was tested in different intervals (up to 60 days), in a dark and humid environment. The ISO 62 standard (plastics-determination of water absorption) was used as a reference for this experiment [38]. The structure of the bio-sequins after exposure to darkness and humidity conditions is summarized in Figure 5.

As observed in Figure 5, the biobased material remained intact and showed no signs of deterioration, preserving its structural integrity even in humid storage conditions. Reduced moisture is preferred as it indicates that the bio-sequin resists water penetration, preventing bacterial and fungi growth. These findings also support the durability of the bio-sequin, further confirming its potential applicability in clothing.

2.1.5. Resistance to UV-A Radiation Exposure

As constant exposure to sunlight can compromise the quality and properties of a material, the resistance of bio-sequins to UV-A exposure was evaluated. In this test, changes in the biobased materials' appearance or structure are observed. Resistance to

UV-A ensures that bio-sequins will preserve their original shine, color, and shape in sunny or outdoor environments. As such, clothes or garments decorated with bio-sequins could be used in such conditions without deteriorating. The tests were performed in accordance to ASTM D4329 [39] recommendations (standard practice for UV-exposure for plastics). Figure 6 shows a bio-sequin after 7 days of exposure to UV-A radiation.

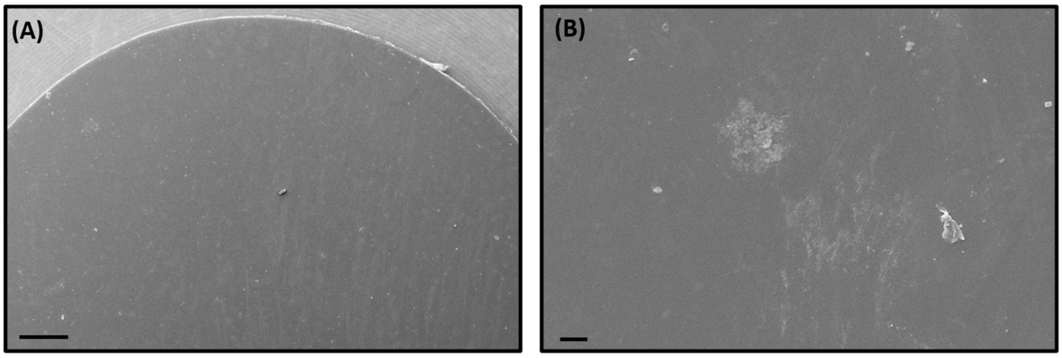

Figure 5. SEM micrographs of BS after 60 days of exposure to darkness and humidity conditions. Scale bars of 1 mm (**A**) and 2 µm (**B**).

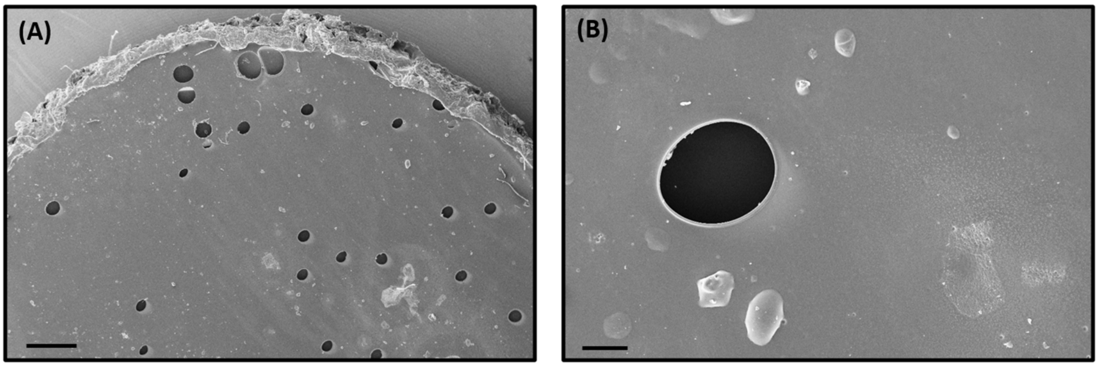

Figure 6. SEM micrographs of the BS after 7 days of exposure to UV-A radiation. Scale bars of 1 mm (**A**) and 100 µm (**B**).

After 7 days of exposure to UV-A, the appearance of pores and slight deformations in the edges of the bio-sequins were recorded probably due to the evaporation of occluded solvents. Despite that, no significant changes in their physical properties were observed. The results demonstrate that the bio-sequins exhibit resistance to the effects of sunlight. Resistance to UV radiation is relevant for clothing that is constantly exposed to outdoor environments. However, future research will evaluate measures to prevent or minimize deformation, further ensuring the long-term durability of the biobased material.

2.1.6. Contact Angle

Contact angle tests were performed to evaluate the hydrophobic properties of the material. This test was conducted by depositing a water droplet on the material's surface. Then, the contact angle formed between the droplet and the surface was measured. Contact angle experiments provide crucial information related to the material's ability to repel or absorb water. If the contact angle is close to 180°, the material presents hydrophobic behavior, thus having low water affinity and repelling it [40]. On the other hand, if the contact angle is close to 0°, the material is hydrophilic, featuring a high capacity for water absorption. Figure 7 describes the contact angle experiment conducted on the bio-sequins.

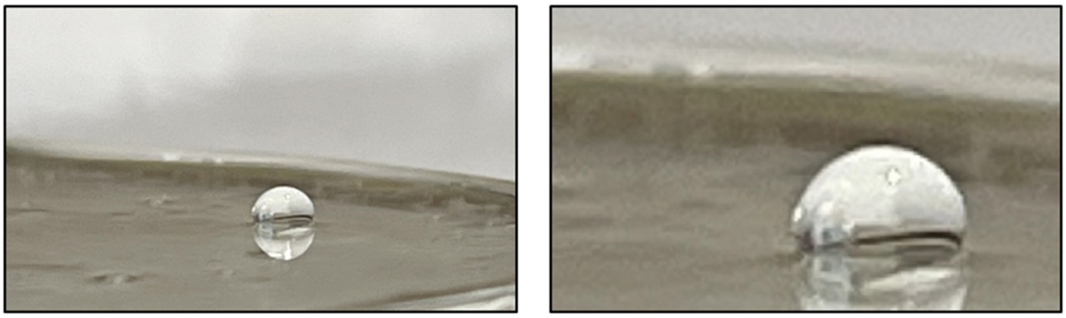

Figure 7. Contact angle test performed on the BS at different magnifications.

As observed from Figure 7, the contact angle test (73.9° ± 7.75°) revealed that the bio-sequin falls within an intermediate range between hydrophilic and hydrophobic behavior. With a contact angle of 73.9°, the bio-based sequin presents mild hydrophobic tendencies [41,42], meaning it does not completely repel water, but it also does not allow it to spread as much as a highly hydrophilic material (contact angle below 30°). This makes the biobased material suitable for applications where resistance to water is required, or for activities where humidity is prominent.

2.2. Precipitation of Guanine Crystals (GC) from Salmon Scales

The extraction of GC from salmon scales was evaluated using centrifugation and stirring methods. Both methods are summarized on Figure 8A–F, respectively.

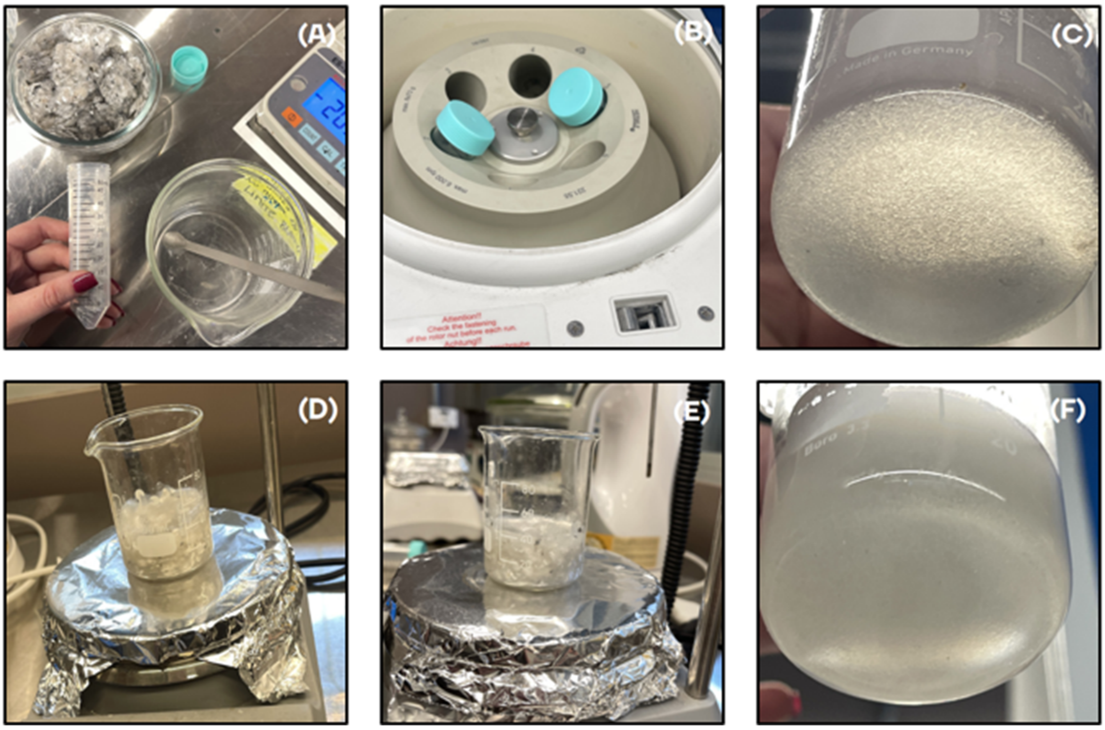

Figure 8. Guanine crystals obtained from centrifugation (**A**–**C**) and stirring (**D**–**F**) methods.

The centrifugation procedure, using acetone as solvent, allowed the rapid precipitation of GC with remarkable brightness and uniform size distribution (Figure 9A). Further, the yield obtained in the centrifugation method was higher than that of the stirring method (Figure 9B). The centrifugation (Figure 9C) and stirring methods (Figure 9D), using ethanol as solvent, also promoted the formation of crystals. These crystals featured considerable brightness, but low size uniformity and agglomerates were observed. As such, the effectiveness of both methods using ethanol must be evaluated or followed by sieving processes.

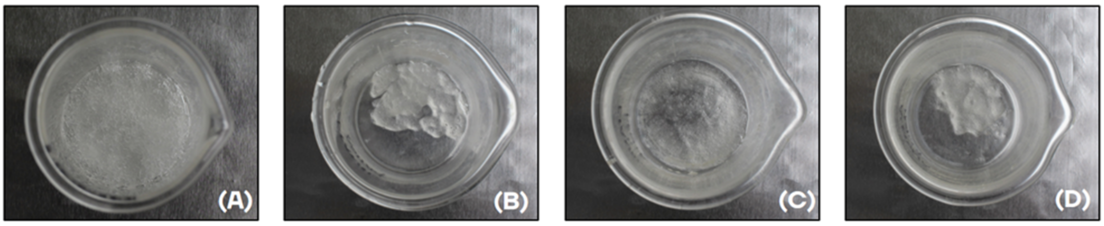

Figure 9. Guanine crystals obtained using acetone (**A**,**B**) and ethanol (**C**,**D**) as solvents.

The choice of acetone and ethanol as solvents is sustainable as they can be reused through fractional distillation, providing an advantage over harmful solvents such as ammonia or strong acids. However, safety precautions, such as the use of gloves and mask, must be implemented when handling and drying the GC.

2.3. Decoration of the Bio-Sequin with GC

The decoration of the bio-sequin with GC was evaluated using two different procedures. The first method involved adding the crystals directly to the mixture described in the methodology. This resulted in the partial dissolution of the crystals, compromising their properties, especially their structure and brightness. The second method consisted in pouring the crystals into the mixture at room temperature, which proved to be more successful. This route allowed the deposition of crystals in the bio-sequin surface, without compromising their signature brightness. Further, a homogeneous distribution of the crystals in the bio-sequin's surface was also observed. This strategy ensured the deposition of the guanine crystals without compromising the structural integrity of the bio-sequin, as well as the quality and visual appeal of the final product. The obtained bio-sequins decorated with GC are illustrated in Figure 10.

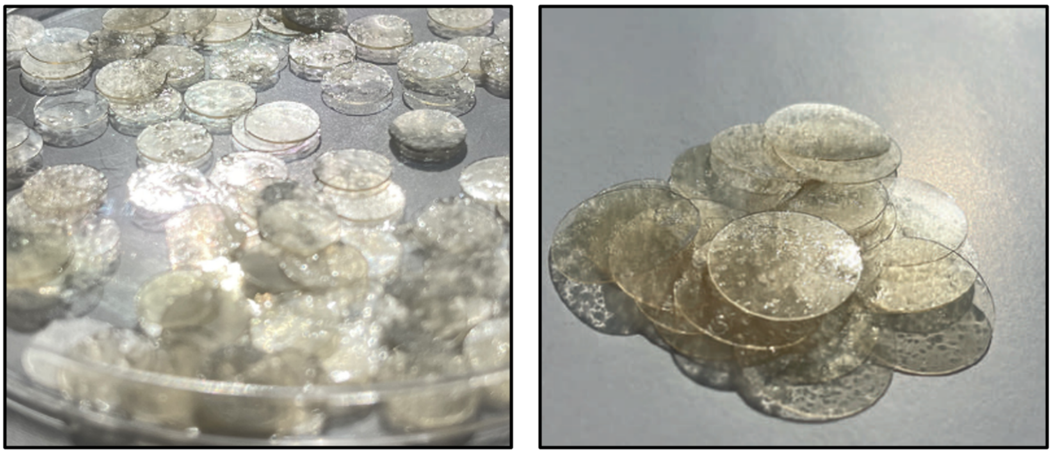

Figure 10. Bio-sequins decorated with GC shown at different magnifications.

2.4. Characterization of the Bio-Sequins Decorated with GC

2.4.1. Scanning Electron Microscopy of the Bio-Sequins Decorated with GC

The deposition of the bio-sequins decorated with GC was ascertained through SEM micrographs. Figure 11 illustrates the SEM and EDS characterization of pristine GC and the bio-sequin decorated with GC.

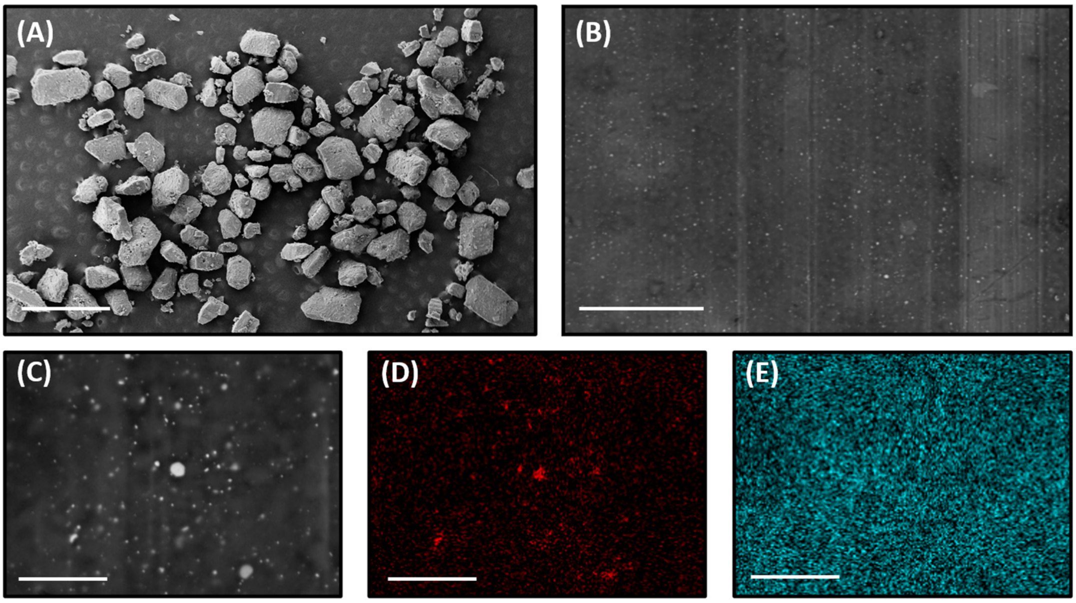

Figure 11. SEM micrographs of GC (**A**, scale bar of 20 µm) and the bio-sequins decorated with GC (**B**, scale bar of 1 mm); area of BS-GC chosen for EDS mapping (**C**, scale bar of 50 µm); EDS mapping (**D**,**E**) of BS-GC, scale bar of 50 µm.

SEM reveals that GC (Figure 11A) exhibit a well-defined, angular morphology. The individual GC appear to have irregular, sharp edges and range in size, indicating a diverse crystal size distribution. This is characteristic of guanine, which tends to form plate-like crystals due to its structure [31,43]. The crystals appear to aggregate, with spaces between them. The gaps suggests that the GC are not densely packed, allowing for interactions with surfaces when incorporated into the bio-sequins. The surface of the bio-sequins in Figure 11B shows that the surface retains its smooth features after deposition of GC, suggesting that the crystals are dispersed over the bio-sequin surface, which could explain the improvement in the thermal stability of the material, as observed in the TGA characterization. The deposition of GC could contribute to improved mechanical properties, which are desirable in functional bio-based materials. Furthermore, the EDS mapping of the BS-GC sample (Figure 11C) detected the presence of C (Figure 11D) and N (Figure 11E), which can be attributed to the amine and amide functional groups inherent to protein structures, the pyrimidine-imidazole ring system of guanine, and peptide bonds.

2.4.2. X-Ray Powder Diffraction Characterization of the Bio-Sequins Decorated with GC

The structural analysis of the bio-sequins decorated with guanine crystals was conducted using X-ray powder diffraction (XRPD). Measurements were taken over a 2θ range of 2° to 80°. As shown in Figure 12, the diffraction patterns of the crystalline material revealed sharp peaks at 10.49°, 17.45°, 25.27°, 28.07°, 30.03°, 31.77°, 32.81°, and 43.35°. Previous studies have shown that biogenic GC, which are naturally occurring in fish skin, can exhibit different XRPD patterns compared to their synthetic counterparts [43], owing to factors such as the presence of other purine metabolites, the orientation of crystals in biological tissues, and the methods of extraction and preparation [32,33]. XRD patterns of GC can also be influenced by the state in which the material is analyzed. Drying the GC prior to analysis could have contributed to the slight differences observed, This aligns with findings from other studies showing that dried GC tend to exhibit distinct crystalline arrangements compared to hydrated GC [44].

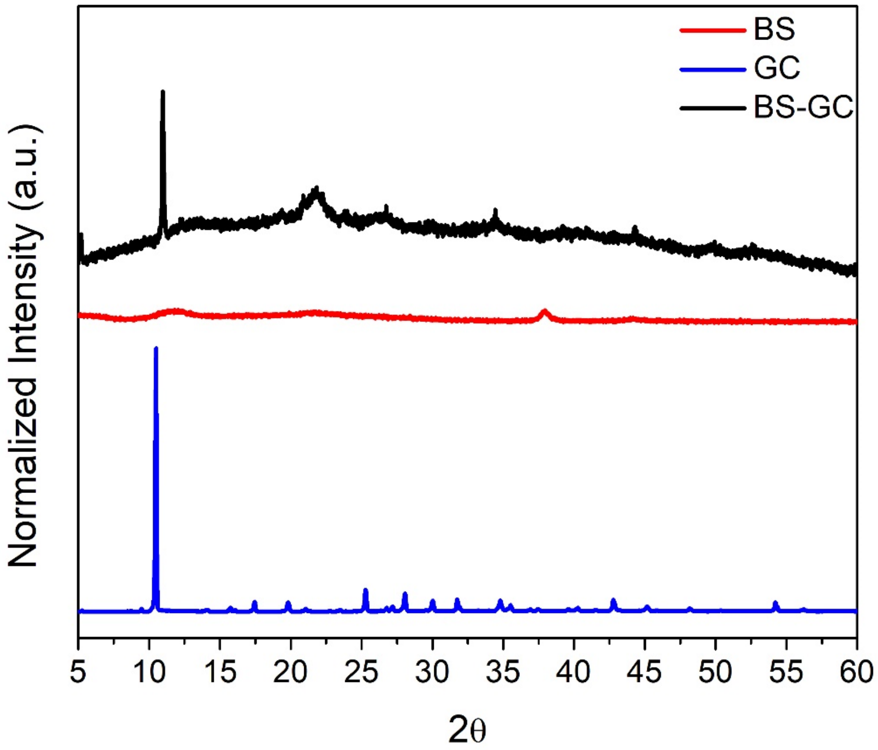

Figure 12. XRPD characterization of BS, GC, and BS-GC.

In contrast, the XRPD pattern of the BS shows broad features, indicative of an amorphous or poorly crystalline structure. The absence of sharp diffraction peaks suggests that BS are composed of predominantly amorphous materials, likely due to the organic nature of their components, such as collagen and glycerin. Organic polymers and biopolymers, like those present in the BS, tend to form amorphous or semi-crystalline structures, which do not produce distinct diffraction peaks in XRPD patterns [45,46]. Moreover, the XRPD pattern of BS-GC shows a combination of the broad, weak features of the BS and the sharp, intense peaks corresponding to GC. While the ordered structure of GC predominates in the diffraction pattern, the overall intensity of these peaks is lower compared to pristine GC, likely ascribed to the immobilization of guanine within the amorphous BS matrix. This observation implies that GC retain their integrity when incorporated into the BS. Furthermore, the BS contribute primarily to the amorphous component of the material.

2.4.3. Thermogravimetric Analysis of the Bio-Sequins Decorated with GC

The bio-sequins, GC, and the bio-sequins decorated with GC were further characterized through TGA. As shown in Figure 13, the degradation peak at 250 °C in the bio-sequin corresponds to the degradation of organic components, such as proteins and small molecules like glycerin. Proteins like collagen or keratin, which are common in animal-based materials, degrade within the temperature range of 200 °C to 300 °C [47,48]. Since the biobased sequins are derived from salmon skin waste, the peak at 250 °C can be attributed to the denaturation and breakdown of protein structures, such as peptide bonds [49]. Part of this peak could also be associated with the volatilization and decomposition of glycerin. Conversely, the bio-sequins decorated with GC exhibit a main degradation peak at 350 °C, which is likely due to the thermal decomposition of more stable organic structures, such as guanine and crosslinked protein residues [50]. Denatured protein structures typically degrade at higher temperatures, within the range of 300 °C to 400 °C. a main degradation peak was observed at 350 °C. The thermal event can be attributed to the protein content and glycerin in the material, as well as possibly the GC embedded onto the bio-sequins. This is consistent with the thermal event observed in the GC profile, which starts to degrade at over 300 °C. GC, as a crystalline material, exhibits higher thermal stability compared to organic polymers like proteins. The sharper slope compared to the bio-sequins suggest that GC experiences a more uniform thermal decomposition, aligning with the single-phase nature of GC. Bio-sequins decorated with GC benefit from the incorporation of the more thermally stable GC, shifting their main degradation event to around 350 °C. This suggests that the GC decoration not only increases the overall stability of the material, but might also contribute to the formation of a composite that resists degradation until higher temperatures.

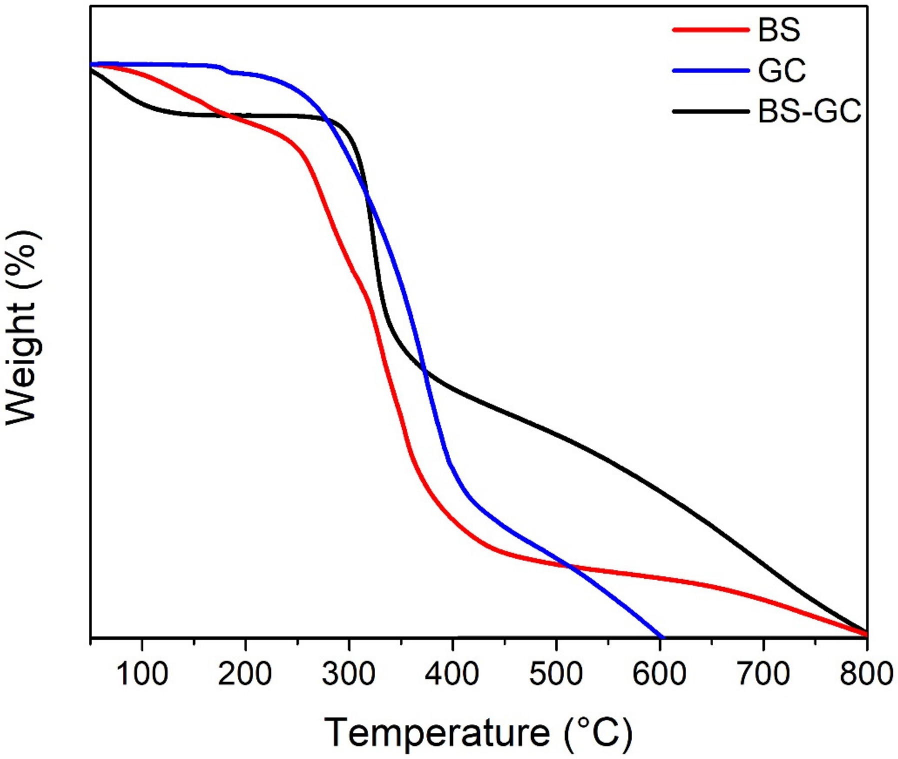

Figure 13. TGA characterization of BS, GC, and BS-GC.

2.4.4. Fourier-Transform Infrared Spectroscopy Characterization of the Bio-Sequins Decorated with GC

After decorating the bio-sequins with guanine crystals, FT-IR spectroscopy was employed to investigate the chemical interactions between the components and the structural integrity of the materials under UV-A exposure (Figure 14). The spectra exhibit characteristic absorption bands that can be attributed to various functional groups present in the samples.

The FT-IR spectra revealed a broad band with relative intensity at 3200–3400 cm^{-1}, attributed to O-H and N-H stretching vibrations. These strong, broad peaks correspond to O-H groups from gelatin, glycerin, and water content, while the N-H stretching from amide groups in gelatin and guanine likely also contributes to this peak. Additionally, bands in the 2800–3000 cm^{-1} range were observed, corresponding to C-H stretching in methylene and methyl groups of organic compounds. The peak at 1650 cm^{-1} is associated with the amide C=O stretching band from peptide bonds in proteins like gelatin and collagen [51,52]. This prominent peak in all samples indicates the preserved integrity of protein structures, even after UV exposure. A band at 1550 cm^{-1}, corresponding to N-H bending, was also observed in all samples, further suggesting that the protein structure remained unchanged. The relative intensity of this peak does not change substantially between the pristine BS-GC and UV-exposed BS-GC, suggesting minimal degradation of the amide bonds after UV exposure. Additionally, peaks around 1300 cm^{-1} are attributed to C-N stretching from the amide groups in the guanine ring [53]. These peaks are most prominent in the GC spectrum, and can also be observed in the BS-GC material, confirming the successful incorporation of GC into the BS. It is worth noting that after exposure to UV-A light for 7 days, there are no major shifts in the main functional group regions (3200–3400 cm^{-1}, 1650 cm^{-1}, and 1550 cm^{-1}), suggesting that the BS material, especially the protein backbone and guanine, retain their structural integrity. Moreover, the slight broadening of O-H and N-H bands after UV-A exposure could indicate increased moisture absorption or conformational changes after irradiation.

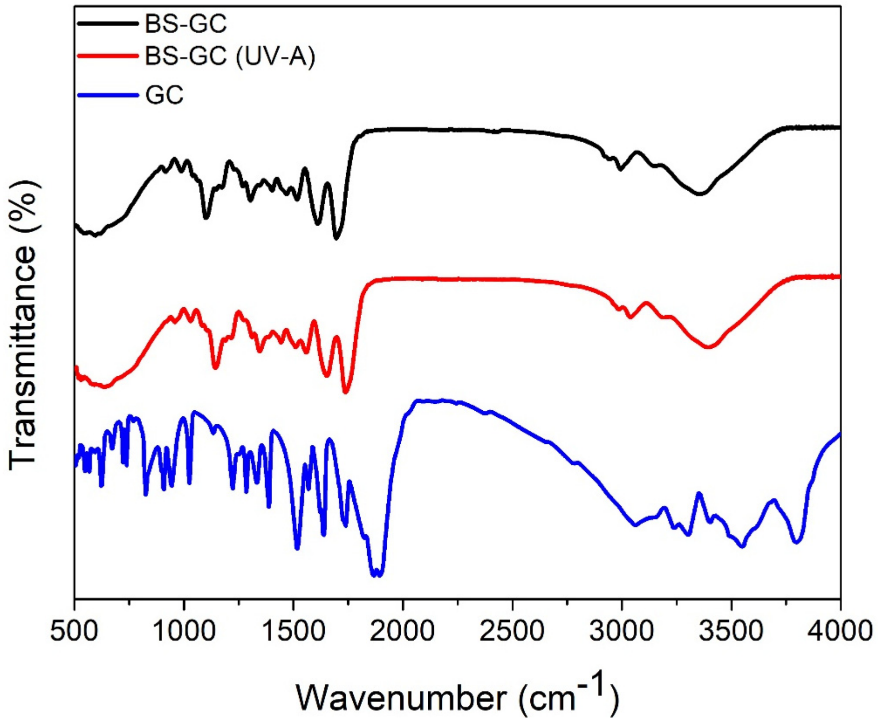

Figure 14. FT-IR spectra of GC, BS-GC, and BS-GC before and after exposure to UV-A radiation.

2.4.5. Mechanical Properties of the Bio-Sequins Decorated with GC

The mechanical properties of bio-sequins decorated with GC, before and after exposure to moisture, UV-A radiation, and physiological temperature were also evaluated. The force-deformation curves are summarized in Figure 15.

The experimental results show that the mechanical behavior of the bio-based sequins is influenced by environmental exposure conditions. Even though all samples display a limit for elastic transition at 2.5 MPa, the increased plastic deformation observed in bio-sequins exposed to moisture can be attributed to the presence of gelatin, a hydrophilic protein known to interact strongly with water. In the presence of moisture, gelatin likely undergoes plasticization, which could lead to a reorganization of the bio-sequin matrix, with water facilitating molecular mobility [54,55]. As a result, the bio-sequin becomes more compliant, deforming under lower stress but retaining its strength after deformation. This suggests that moisture enhances flexibility without compromising the material's integrity. Notably, the bio-sequin exposed to UV-A radiation exhibits a significant increase in mechanical strength, likely due to UV-A-induced crosslinking within the material. Indeed, UV light has been reported to induce crosslinking in protein-based materials like gelatin, enhancing mechanical strength and stiffness [56,57]. Crosslinking forms additional covalent bonds between amino acid chains, increasing the material's resistance to plastic deformation. The bio-sequin exposed to physiological temperatures (37–42 °C) also shows slight strengthening. Gelatin is known to exhibit thermal behavior, undergoing a gel-to-sol transition when heated, which may result in a reorganization of the biopolymer network, improving mechanical properties [58,59]. Environmental conditions likely alter the internal structure of the bio-sequin, making its polymer matrix denser and more tightly packed, contributing to the observed increase in tensile strength. Guanine crystals (GC) may also play a role in the changes in tensile strength. GC are rigid, hard materials that could reinforce the bio-sequin matrix. As the GC are dispersed throughout the matrix, they form a composite, where the GC act as a stiff phase, helping to distribute stress more evenly and preventing deformation. UV exposure might promote crosslinking between the protein matrix and the GC, strengthening the interface. Similarly, moisture could enhance the interaction between the protein matrix and GC, as gelatin swells in the presence of water, improving stress distribution around the embedded GC and increasing the material's flexibility and toughness [60,61]. Temperature exposure could also induce slight surface alterations in the bio-sequin, improving adhesion between the GC and the protein matrix, which may explain the increased strength after exposure to physiological temperatures.

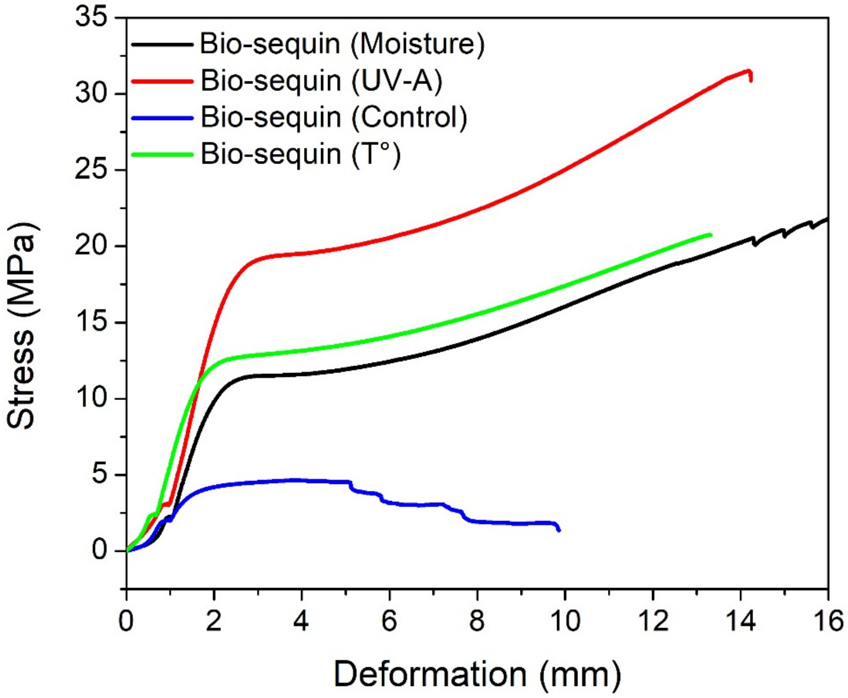

Figure 15. Force-deformation curve of the BS-GC before and after exposure to moisture, UV-A radiation, and temperature (37–42 °C).

3. Potential Applications of the Bio-Sequins Manufactured from Salmon Waste

The potential application of bio-sequins manufactured from salmon waste offers a wide spectrum of possibilities, primarly in sustainable design. The developed bio-sequins are biodegradable, versatile, and compile with fashion standards, while also presenting an ecological approach by upcycling waste. Beyond their functional reinforcement role, GC also provide an aesthetic role, contributing to the iridescence of the bio-sequins. As illustrated in Figure 16, the use of our proposed biobased sequins can be envisioned for clothing, accessories, textiles, and decoration, presenting as an ethical and ecological alternative to conventional petroleum-based sequins.

In terms of visual enhancement, Figure 17 illustrates the impact of GC incorporation on the aesthetic properties of BS.

Figure 17A shows the BS without GC, demonstrating a surface with minimal reflective properties. In contrast, Figure 17B highlight the enhanced brightness and reflective qualities introduced by the addition of GC, showcasing that its crystalline structure produces a pearlescent effect. Finally, the incorporation of a vegetal pigment, as shown in Figure 17C, further amplifies the decorative potential of the biobased sequin, enhancing both color vibrancy and light reflectance. The improved brightness and decorative flexibility offered by BS-GC pave the way for their use in couture, ornaments, textiles, and interior design, aligning with the growing demand for sustainable materials. The use of these bio-sequins in the textile industry is not only sophisticated and innovative, but also promotes the integration of environmental responsibility and circular economy, addressing the issue of waste management. This biobased material represents a promising step towards sustainability in the textile industry, demonstrating that innovation can converge with environmental awareness.

Figure 16. Potential applications of BS-GC manufactured from salmon waste.

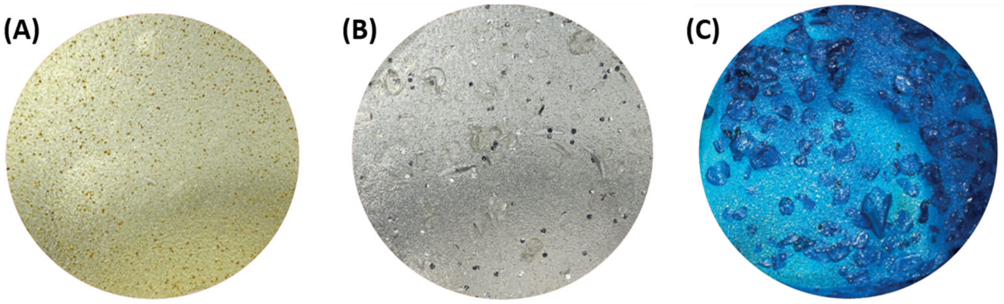

Figure 17. Visual enhancement of BS (**A**) after incorporation of GC (**B**) and GC-vegetal pigments (**C**).

4. Experiments and Methods

4.1. Materials

Sodium Bicarbonate, $NaHCO_3$, 99%, 84.01 g/mol; glycerin, $C_3H_8O_3$, 99%, 92.09 g/mol; acetic acid, $C_2H_4O_2$, 99%, 60.05 g/mol; ethanol, C_2H_5OH, 99.5%, 46.07 g/mol; acetone, CH_3COCH_3, 99.5%, 58.08 g/mol were purchased from Sigma Aldrich (St Louis, MO, USA) and used as received with no further purification. Milli-Q water (0.05 μS cm^{-1}; 18 MΩ cm) was obtained through a Millipore Direct 5 system and used to prepare the solutions for this study.

4.2. Extraction of the Raw Material from Salmon Scales

A thorough investigation was carried out to identify sources of waste from the salmon industry. The raw material used in this work was provided by two main locations: the fish market and the supermarket fishmongers, where a significant amount of the waste material was generated. It is worth noting that the raw material was collected weekly, allowing to gather 30 salmon skins in total.

4.3. Pre-Treatment of the Raw Material

Once acquired, the salmon skins underwent a meticulous pre-treatment process. These processes were performed to ensure the quality and purity of the obtained salmon scales, which constituted the main component of the biobased materials. First, the skins were classified based on their size. Subsequently, a cleaning stage was performed using water. All remnants of biological matter that may remain in the salmon skin, including bones, fins, and cartilage, were removed. Subsequently, a scaler or a sharp knife was used in an extraction procedure on the salmon skin. Extraction of the salmon scales was performed from the tail to the head in a perpendicular motion on the skin. This process required up to 6 min depending on the size of the skin. As the scales were extracted, they were moisturized with water to prevent adhesion. All collected scales were cleaned up with abundant distilled water. To obtain a higher-purity raw material, the scales were treated with 14.7 g of $NaHCO_3$ in 1 L of distilled water for 3 h. Then, the scales were strained and left to settle for 12 h to remove residual water. The scales were dried with absorbent paper before being placed in a desiccator for 3 h. After the drying process, the scales were grinded and sieved with a 75 µm mesh to separate the scales from agglomerations or impurities, while also providing size uniformity to the raw material. Depending on the desired application, the resulting scales can be used either raw or dehydrated.

4.4. Manufacture of the Bio-Sequins

The pre-treated salmon scales were blended and homogenized with distilled water, glycerin, gelatin, and white vinegar to obtain the biobased sequins. First, 120 mL of distilled water were added to a pot. 24 g of gelatin, which acts as a binding material and hardening component [16], incorporated and homogenized in the distilled water at room temperature. Moreover, the development of the bio-sequins involved the incorporation of a plasticizer, in this case, glycerin [62]. The temperature of the mixture was increased to 60 °C and then a fixed amount of glycerin (6 mL) was added. To hinder microbial and fungi growth, white vinegar [63] (5 mL) was added to the mixture as the last ingredient. This component is crucial to extend the lifespan of the biobased material. Once a homogeneous consistency was attained, the mixture was poured into Petri dishes and allowed to dry in a desiccator for 8 h.

4.5. Extraction of Guanine Crystals from Salmon Scales

Guanine crystals were extracted from the salmon scales using two different procedures. The selected methods were based on previously reported articles [33,64] with modifications. As guanine extraction sequences often occur in HCl or NH_3, we focused on developing greener alternatives that do not involve the use of such hazardous solvents. The precipitation of GC from salmon scales was performed using appropriate solvents, such as acetic acid, ethanol, and acetone. While acetone, ethanol, and acetic acid are not entirely innocuous, they are less harmful than traditional solvents [65–67] and can be recycled through fractional distillation [68,69], extending their lifespan and reinforcing the sustainable approach in all the stages of manufacture. The first step involved an initial extraction with acetic acid to break down the proteins and release guanine. Then, the solution was neutralized using $NaHCO_3$ to promote protein precipitation. Next, the separation of guanine crystals was performed through two different procedures: centrifugation, whereas the second method consisted on the formation of GC under constant stirring. The centrifugation method was performed using 50.3 g of salmon scales, which were added to 500 mL of acetic acid (5% v/v) and allowed to settle for 2 h. After neutralization with $NaHCO_3$ and filtration to remove solid proteins residues, a solution of ethanol (70:30 v/v%) or acetone (70:30 v/v%) were added to the filtrate containing guanine in a Falcon conical tube. After settling for 20 min, the salmon scale/ethanol mixture was centrifuged at 6000 rpm for 1 h. The salmon scales were separated from the supernatant and the centrifugation was repeated twice under the same conditions. After 48 h, the guanine crystal suspensions were observed to settle, separated from the solution, and dried in a desiccator for 24 h. The agitation method was evaluated as another procedure to extract guanine crystals, which was performed as follows: 50.3 g of salmon scales were added to a beaker containing a solution of acetic acid (5% v/v) and allowed to settle for 2 h. After gradually adding Na_2CO_3 and filtrating to remove solid protein residues, ethanol (70:30 v/v%) or acetone (70:30 v/v%) were added in increments of 50 mL under stirring. The mixture was left under constant stirring for 60 min. After 48 h, guanine crystals were settled, filtered, and rinsed with cold ethanol or acetone. Finally, the obtained crystals were dried in a desiccator for 24 h.

4.6. Characterization of the Developed Bio-Sequins

To understand the biobased materials properties and their suitability for use in specific applications, the physical and mechanical characterization of the biobased materials was performed.

4.6.1. Resistance to Physiological Temperature

Resistance to physiological temperature (37–42 °C) was measured to simulate body temperature and determine the potential application of the BS in clothing. The biobased sequin samples were cut into consistent pieces. Then, the samples were placed inside an incubator and set at a range between 37–42 °C to simulate body temperature. The test was run over 24 h. to simulate prolonged wear conditions. The relative humidity to replicate skin-like conditions was controlled (60% RH). The samples were monitored at 2 h. intervals to assess any visible changes, such as color alteration, flexibility, or degradation.

4.6.2. Resistance to Moisture Under Storage Conditions

To assess resistance to moisture of the BS, the experiments were based on ISO 62: plastics-determination of water absorption. This standard is used to evaluate how much moisture a material absorbs under specified conditions, which is crucial for textile applications. Briefly, the samples were cut in consistent dimensions, and dried thoroughly in an oven at 60 °C for 24 h. After drying, the samples were cooled in a desiccator, and weighed to record their initial weight. Based on ISO 62 recommendations, resistance to moisture was monitored up to two months. After completing the test, the biobased sequins were analyzed to assess their durability under humid storage conditions.

4.6.3. Resistance to UV-Rays Exposure

The resistance to UV-ray exposure of the BS was evaluated according to ASTM D4329 standards (standard practice for UV exposure for plastics). Briefly, the samples were prepared in uniform size and thickness. Properties such as color, gloss, and elongation were chosen as baseline parameters before measurements. Then, the biobased sequins were mounted in a chamber and exposed to UV using a UVA-340 lamp, which simulates sunlight exposure (295 nm). A total exposure time of 200 h. was chosen, with exposure cycles of 8 h. The bio-sequin condition, such as changes in color, surface gloss, or cracking, were assessed after exposure.

4.6.4. Contact Angle Test

A contact angle test was performed to evaluate the wetting properties of the BS by measuring the angle between a droplet and the surface of the material. This analysis gives insight into the hydrophobicity or hydrophilicity of the BS. The biobased material was placed on a flat-horizontal surface. Then, 10 µL droplet of distilled water was deposited on the material surface, ensuring that the droplet remained stable, without spreading excessively or evaporating. Then, a side-view image of the liquid droplet was acquired. The contact angle (θ) was determined by the angle formed between the tangent to the liquid droplet at the contact point and the surface of the material, which was obtained using ImageJ 1.53 software.

4.6.5. X-Ray Powder Diffraction

The X-Ray diffraction technique was used to stablish the crystalline structures of the BS, the GC, and the bio-based sequins decorated with GC (BS-GC). The analysis was performed by using X-ray powder diffraction (XRPD) with a Bruker D5000 diffractometer. The experiment was performed with a Cu Kα radiation source. A Ni filter was used, and the data was gathered within an angular range of 2–80° 2θ and scan speed of 0.05°/s.

4.6.6. Thermogravimetric Analysis

Thermogravimetric Analysis (TGA) of BS, the GC, and the BS-GC samples was performed on a TGA-4000 Pyris within a 25–800 °C temperature range to stablish the limits for temperature degradation of the materials. The samples were placed on an aluminum pan, and measurements were conducted under N_2 atmosphere.

4.6.7. Scanning Electron Microscopy and Energy Dispersive Spectroscopy

The morphology and elemental analysis of the biobased materials and deposition of GC were examined using a Zeiss LEO-Supra 35-VP scanning electron microscope (SEM) operating at 20 kV. All samples were deposited directly onto carbon tapes, and gold coating was performed using a magneton sputtering. The deposition of GC onto the organic bio-sequins was also assessed through energy dispersive spectroscopy (EDS) and elemental mapping.

4.6.8. Fourier Transform Infrared Spectroscopy

Fourier Transform Infrared (FT-IR) spectra of the biobased sequins were acquired using a Nicolet iS5 equipment, within a range of 500–4000 cm^{-1}, averaged over 32 scans and a resolution of 0.2 cm^{-1}. This technique allowed to assess any molecular change of the proposed materials after being subjected to different treatments.

4.6.9. Mechanical Strength Tests

The mechanical properties of the BS and BS-GC were determined using a universal tensile testing machine (Zwick/Roell Zoo5, Ulm, Germany). Tests were performed on 5 cm × 1 cm strips, and the nominal deformation was determined at a traverse speed of 5 mm/s in a displacement-controlled manner. This technique allowed to assess the viscoelastic behavior of the proposed materials after being subjected to different treatments.

4.6.10. Biodegradability in Soil

In accordance to the sustainable focus of this work, biodegradability tests in soil were also assessed. The BS were cut into uniform pieces. Then, the samples were dried and weighed before degradation. The sample was mixed with soil (2–3 parts of soil to 1 part of sample).

The soil temperature and moisture content were measured periodically. According to ISO 17556 standards [70], the biobased material can be considered biodegradable in soil if there is at least 90% degradation within 6 months.

5. Conclusions and Final Remarks

This study demonstrates the feasibility of producing bio-sequins from salmon skin waste, incorporating guanine crystals for decorative purposes, and provides insights into their potential as sustainable alternatives to conventional plastic sequins. The mechanical properties and physicochemical characteristics of the resulting biobased materials confirm the effectiveness of the proposed methodologies, showcasing innovations in both material development and sustainable extraction processes. Our hypothesis that salmon skin, a by-product of the salmon industry, could be successfully transformed into biodegradable sequins was supported by the results. The extraction of guanine crystals using less hazardous solvents like acetic acid and acetone, coupled with centrifugation, proved to be an effective and environmentally friendly method. This marks a significant departure from traditional extraction methods involving hazardous chemicals like hydrochloric acid or ammonia, aligning with the global shift towards greener, safer processes. Furthermore, biodegradability test results are particularly promising, revealing that the bio-sequins decompose rapidly in soil, making them a sustainable option for reducing plastic waste in the environment. This high biodegradability can be attributed to the organic nature of the bio-sequins, which accelerates its degradation process. While this characteristic enhances biodegradability, it will be important to monitor the biobased sequin's performance in various environments to prevent any premature degradation during use. Further validation on the biodegradability of the GC-decorated biosequins are needed to confirm their environmental impact across different conditions.

The incorporation of guanine crystals enhanced the aesthetic appeal of the bio-sequins, although further optimization is necessary to achieve uniform crystal distribution on the material's surface. The use of a strainer or sieve during application would likely resolve this issue, ensuring more even coverage and better performance. Compared to other biobased plastics, the bio-sequins developed in this study offer several key improvements. Their production from fish waste not only addresses the environmental concerns associated with synthetic polymers but also promotes waste valorization and circular economy principles. Additionally, the drying process was optimized at 35 °C, allowing for better preservation of the mechanical and physical properties of the bio-sequins, resulting in a smooth, uniform texture that facilitates easier handling during manufacturing stages such as drilling and finishing. These features set the bio-sequins apart from other biobased plastics, which often struggle to balance mechanical strength with biodegradability. However, this study also presents certain limitations. One practical concern is the potential vulnerability of the bio-sequins to damage during washing and tumbling, particularly at temperatures over 40 °C. The gelatin matrix, which contributes to the material's biodegradability, may soften under such conditions, leading to a possible loss of structural integrity. Further testing is necessary to evaluate the durability of bio-sequins when exposed to laundering processes, and future research could focus on enhancing their resistance to washing while maintaining biodegradability. The mechanical properties of the bio-sequins, while improved compared to conventional sequins, may still need enhancement for broader applications, especially in areas requiring high durability. Additionally, the scale-up of the production process will require further refinement, particularly in terms of guanine crystal distribution and overall manufacturing efficiency. The compatibility of the bio-sequins with various textile substrates has yet to be fully explored, and further testing is needed to evaluate their long-term performance under environmental conditions such as UV exposure, moisture, and wear and tear. Looking ahead, future work should focus on optimizing the integration of guanine crystals into the bio-sequin matrix, exploring automated or large-scale processes to ensure uniform distribution. Further research is also needed to explore the potential applications of bio-sequins beyond the fashion industry, such as in decorative arts, interior design, and sustainable packaging. Investigating the use of other bio-based or waste-derived materials in combination with these bio-sequins could lead to a new generation of sustainable, multifunctional materials. Moreover, life cycle assessments (LCAs) should be conducted to better quantify the environmental impact of the bio-sequins compared to conventional plastic sequins. Ultimately, the development of these bio-sequins represents a significant step toward more sustainable design practices, offering a promising alternative to traditional, petroleum-based sequins. With continued research and optimization, bio-sequins have the potential to contribute meaningfully to reducing plastic pollution, promoting waste valorization, and supporting the circular economy within the fashion and textile industries.

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