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Article

Physiochemical and Mechanical Evaluations of Electrospun PLA/Microporgonias Undulatus- Gelatin Fibres for Wound Dressing Applications

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Abstract

The poor performance of neat polylactic acid (PLA) and gelatin has driven the development of co-electrospun composites in biomaterials to achieve enhanced functional properties. In this study gelatin extracted from Crocker fish scale was co-electrospun with PLA. The composite fibres were fabricated at 2-17 wt.% gelatin. The electrospun fibres were evaluated via scanning electron microscope (SEM), Fourier transform spectroscopy (FTIR), differential scanning microscopy (DSC), thermogravimetric analysis (TGA), and Tensile test. FTIR analysis of PLA/gelatin fibres showed a peak growth at 1525cm⁻¹ and a shift in the amide III band from 1239 cm⁻¹ to 1192cm⁻¹, indicating hydrogen bonding and chemical interaction between PLA and gelatin. The thermogram of the PLA/gelatin scaffold revealed an enhanced thermal stability with peak thermal stability (324°C) attained at 14 wt.% Gelatin. The DSC further confirms the interaction of PLA (T_g 75 °C) and gelatin (49 °C), forming a single glass transition temperature (T_g) at 70 °C. There was a slight increase in T_g of the composite fibres as the wight fraction of the gelatin increased. The SEM showed a good morphology resembling the native extracellular matrix (ECM) of the body. The ultimate tensile strength and percentage elongation of the fibres declined with increasing gelatin content. The introduction of this gelatin into PLA resulted in improved physiochemical properties of PLA/gelatin fibres due to chemical interaction. Thus, this composite fibre could serve as a potential wound dressing material.

Keywords: PLA; fish scale; thermal stability; wound dressing; gelatin

1. Introduction

Fibrous structures at the nano and microscale can be produced from both natural and synthetic polymers via electrospinning. Electrospinning is an innovative and versatile technique that has been widely employed for fabricating fibrous materials because of its simplicity, continuous fibre production, precise control over fibre size, tunable porosity, and ease of surface functionalization [1].

Electrospun fibres with highly porous microstructures closely mimic the architecture of native extracellular matrices (ECMs), thereby enhancing cellular adhesion, proliferation and differentiation [2]. Consequently, electrospun scaffolds have found numerous applications in the biomedical and healthcare industries, particularly in Tissue engineering (TE) and Regenerative medicine (RM). Tissue engineering seeks to repair or regenerate defective tissues by combining biomaterials, cells, and bioactive factors [3]. Electrospun fibres are also widely explored for drug delivery, bone and cartilage regeneration, wound dressings, and advanced scaffolds for regenerative medicine [4–6].

More than 100 synthetic polymers have been successfully electrospun due to the simplicity and scalability of the process [7,8]. Among them, Poly-lactic acid (PLA) and its copolymer poly (lactic-co-

glycolic acid) (PLGA) are the most widely used in biomedical application. Their popularity is attributed to their biocompatibility, biodegradability, non-toxicity, and mechanical stability [9]. PLA in particular is well recognized as a biodegradable polymer, with diverse applications, including medical implants and scaffold for tissue engineering. Its favourable properties include easy processability, excellent biocompatibility, a bioresorption rate that can be tuned to match tissue healing times, and the generation of non-toxic byproducts upon degradation [5]. However, PLA has limitations such as high hydrophobicity and local acidification during degradation impair cellular attachment, infiltration, and proliferation [10]. Co-spinning PLA with natural polymers, such as gelatin, is a promising strategy to overcome these drawbacks.

Gelatin, a natural polymer derived from collagen, exhibits excellent biocompatibility and biodegradability. It contains bioactive motifs, including arginine-glycine-aspartic acid (RGD) sequences, which promote cell adhesion, proliferation, and differentiation [11]. Nevertheless, gelatin's poor mechanical strength, rapid degradation, and high hydrophilicity limit its use as a standalone scaffold material [12]. When blended with synthetic polymer like PLA, gelatin can enhance biological interactions while PLA provides the necessary mechanical stability and durability [1].

Several studies have investigated PLA/Gelatin composites and reported improved biological and physiochemical properties compared to PLA alone [12,13,15]. Gelatin, which is attractive due to its low cost and wide availability, is however, commercially sourced from Bovine and porcine tissues [16]. Concern over zoonotic diseases such as foot and mouth disease, as well as dietary and religious restrictions have driven the search for alternative gelatin sources. Fish-derived gelatin has emerged as a viable substitute offering comparable biocompatibility with fewer health and ethical concerns [8]. An *et al.* (2010) [8] successfully electrospun composite nanofibers from poly(L-lactide) (PLLA) and gelatin extracted from catfish skin, achieving a scaffold with improved mechanical performance. More recently, gelatin has been extracted from various fish species, demonstrating promising results in biomedical applications [17,18].

In this study, gelatin was extracted from croaker fish scales of Nigerian origin and blended with PLA to fabricate electrospun scaffolds. To the best of our knowledge, electrospun PLA/gelatin composites derived from croaker fish scales have not been extensively reported. This research aligns with United Nations Sustainable Development Goals, particularly SDG 3(Good health and wellbeing), by advancing biocompatible wound dressing materials to support tissue regeneration and improved healthcare outcomes and SDG 12(responsible Consumption and Production),by transforming fish waste into sustainable materials, thereby promoting circular economy practices and reducing environmental impacts from marine by products. This study, therefore, seeks to explore their potential for biomedical applications, particularly in tissue engineering.

2. Materials and Methods

2.1. Materials

Poly(lactic acid) (PLA) with an average molecular weight of 250,000 g/mol was obtained from Nature Works (Suzhou, China). Croaker fish scales were collected from Ojuelegba market, Lagos, Nigeria. Hydrochloric acid (HCl) and Dichloromethane (DCM), 95% purity were purchased from Sigma Alderich; while deionize water, was obtained from Metallurgical and Materials Engineering Laboratory, University of Lagos.

2.2. Methods

2.2.1. Extraction of Gelatin

The extraction of gelatin followed an acid pretreatment process. Approximately 200g of croaker fish scales were thoroughly washed with deionized water to remove surface impurities and oven dried at 70 °C to a constant weight. The dried scales were then immersed in 2.2 M of HCl, at a

scale/solution ratio of 4:10 (w/v) for three hours (3 hrs) to decalcify and remove residual minerals. After decalcification, the samples were repeatedly washed with deionized water until a neutral pH was achieved. The scales were subsequently subjected to water bath extraction at 70 °C for 8 h using a fish scale/water ratio of 3:10 (w/v). The resulting gelatin solution was filtered through Hoffman filter paper (size 5) and freeze-dried for 24 hours to obtain powdered croaker fish gelatin.

2.2.2. Electrospinning of PLA/Gelatin

A 20% (w/v) of PLA solution was prepared by dissolving PLA pellets in dichloromethane (DCM) under magnetic stirring at 300 rpm until homogeneity was attained. Separately, gelatin was dissolved in 10 mL of deionized water at varying concentration (2 -17wt.%). The gelatin solutions were then added to the PLA solution in corresponding weight ratios and stirred for an additional 10 min to ensure uniform mixing. The prepared polymer solutions were loaded into a 10 mL syringe fitted with 21-gauge stainless steel needle. Electrospinning was carried out at a voltage of 15 KV, with the collector distance maintained at 12 cm. The fibres were collected on aluminium foil substrates and subsequently dried to remove residual solvents before characterisation.

2.3. Characterisation of Electrospun PLA/Gelatin Scaffolds

The morphological feature of PLA/gelatin was examined using a scanning microscope (Phenom Eindhoven, Netherlands). The samples were affixed on copper stub and was coated with gold for SEM observation. Functional group analysis via Fourier Transform Infrared Spectroscopy (FTIR) of the electrospun PLA/gelatin were done using Agilent Technologies Cary 630 spectrometer (India). The glass transition temperature (T_g) was observed using Mettler Toledo, DSC 1 star system. 5 mg of the samples were cut prior to the analysis. Thermal history of PLA/gelatin was carried out on TGA(Q500). 2mg of the fibres were heated to 750°C at 10°C/min. The tensile strength analysis of PLA/gelatin fibre was done using Instron Model 313. The electron samples were cut into 50 mm by 10 mm and were firmly held at both ends the samples were deformed until failure occurred.

3. Results and Discussion

3.1. Chemical Analysis of Electrospun PLA and PLA/Gelatin Scaffold

Electrospun neat PLA exhibits a weak asymmetric band of 1740 cm^{-1} (Figure 1) that is attributed to C=O carbonyl stretching, with another absorption band of 1445 cm^{-1} , assigned to -CH stretching, which is approximately within the range reported by earlier studies [19–21]. The band at 1180 cm^{-1} is assigned to the ester group C-O, while 867 cm^{-1} is assigned to C-C stretching of the amorphous phase. Gelatin major peaks were found around 3304 cm^{-1} , 1645 cm^{-1} , 1527 cm^{-1} 1239 cm^{-1} , which are respectively assigned to OH group, C=O bending in amide I, C-H/C-H bending in amide II region, and amide III and 1058 [22]. These peaks were also in the same range, with standard gelatin reported by other researchers [1,23–27]. The PLA/Gelatin electrospun composite showed more of the PLA spectral, with identical peaks irrespective of different weight fraction of gelatin; however, there is a peak growth in the band of 1525 cm^{-1} for the PLA/Gelatin electrospun fibre. The amide I/II absorption band completely disappear as can be seen from the FTIR spectral of the electrospun fibre composite, which shows that PLA completely engulf the gelatin in the matrix. However, there was a slight peak shift in the amide III band (1239 cm^{-1}) in gelatin to 1192 cm^{-1} in the electrospun PLA/Gelatin composite, which established chemical interaction between PLA and gelatin. The peak shift is attributed to intermolecular hydrogen bonding within the functional groups [28]. The shift in peak has led to an enhanced C-H bond strength of electrospun PLA/Gelatin composite, which increased the stiffness and thermal stability of the composite fibre. Furthermore, another growth and broadening of peaks in the electrospun composite was observed, which were negligible in the spectra of gelatin and PLA. This could be attributed to the mechanism of superimposition of similar functional groups, leading

to increment in the amount of the functional group present within the absorption band. The peak continued to grow as the percentage weight of gelatin increased from 2 to 17wt.%.

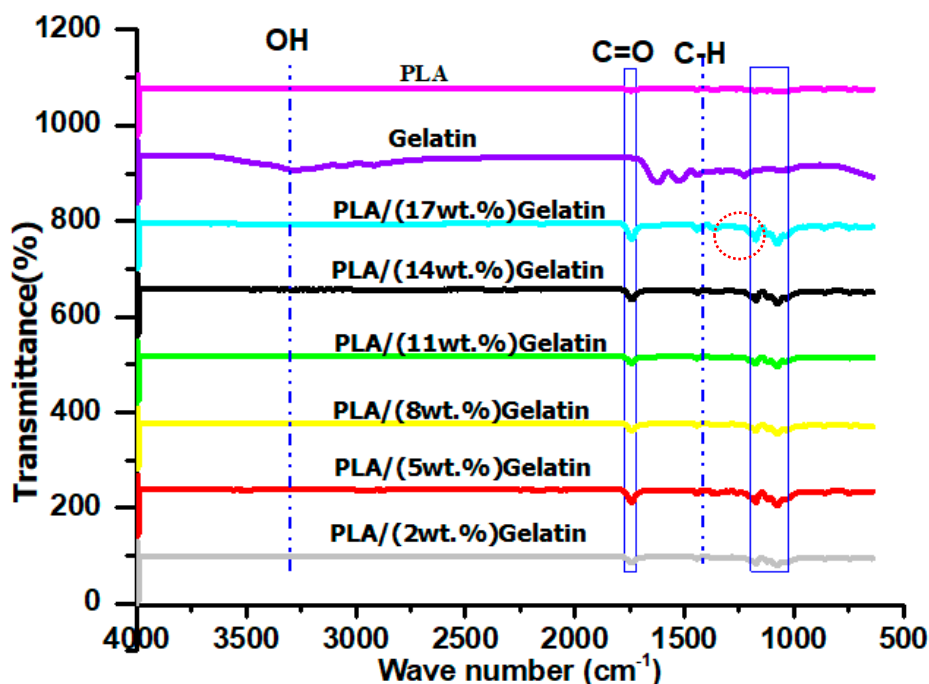


Figure 1. FTIR spectral of electrospun PLA and PLA/Gelatin Composite.

3.2. Thermal Properties of Electrospun PLA and PLA/Gelatin Scaffold

The thermogram of the electrospun PLA, gelatin, and its composites is shown in Figure 2. PLA exhibited a one-step decomposition, which agrees with Branco *et al.* (2018) and Nooeaid *et al.* (2020) reports [21,27]. Polylactide onset temperature is 302°C as seen in Table 1, with a maximum weight loss of 70 wt.% and decomposition temperature of 397 °C. The weight loss is as a result of the thermal decomposition of the electrospun PLA. However, gelatin onset temperature is 313 °C. It exhibits three-step decompositions, as reported by Nooeaid *et al.* (2020)[21]. Gelatin's initial weight loss of 3 wt.% at 170 °C, is attributed to loss of moisture. The second stage is the decomposition of amino acids at 360°C with about 25 wt.% loss and at 480°C decomposition of gelatin network [29]. The electrospun PLA/Gelatin composite fibre exhibited three stage decomposition characteristics of gelatin. The electrospun PLA/gelatin fibre mat becomes more thermally stable, compared to the individual polymer. The improved thermal stability is attributed to better interaction between PLA and Gelatin [30]. It is important to note that gelatin contains functional group such as the hydroxyl group and amine group, which can cross link with PLA molecules. This interaction has the capability to reduce the movement of the polymer chain, thus increasing the thermal stability of the electrospun PLA/Gelatin fibre mat. The improved thermal stability of the electrospun fibre mat varies from 316°C at 8 wt.% to peak of 324 °C at 14 wt.% gelatin. Thereafter there was a decline in the thermal stability to 271°C at 17 wt.%. Figure 3 shows the DTG of the individual polymer and the composite fibre mat. Gelatin showed maximum decomposition temperature at 397 °C, while PLA, PLA/gelatin (2 wt.%), PLA/gelatin (5 wt.%) demonstrated similar maximum decomposition temperature at 415 °C. Similarly, PLA/ gelatin (11 wt.%), PLA/gelatin (8 wt.%) exhibited maximum decomposition temperature of 385 °C. PLA/gelatin (14 wt.%) and PLA/gelatin (17 wt.%) has their decomposition temperature at 440 °C and 470 °C respectively.

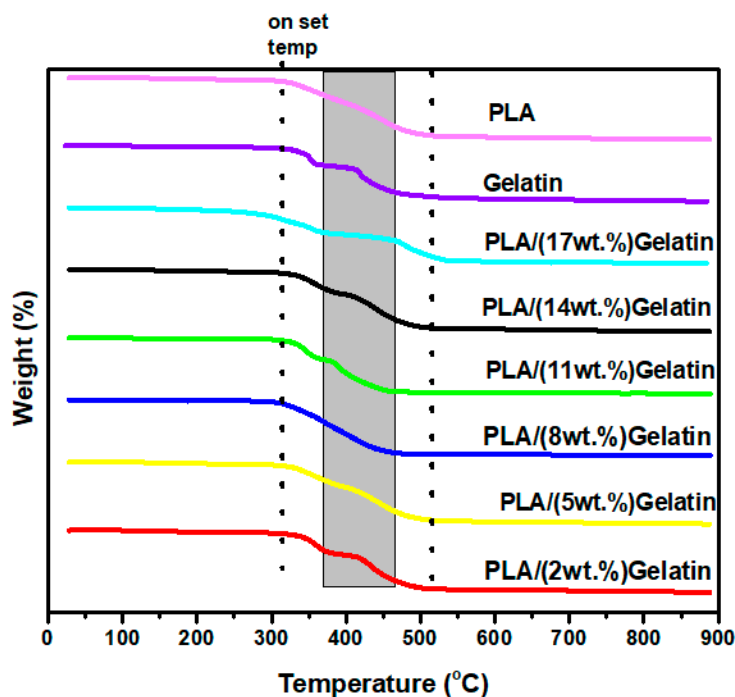


Figure 2. Thermogram of electrospun PLA and PLA/Gelatin Composite.

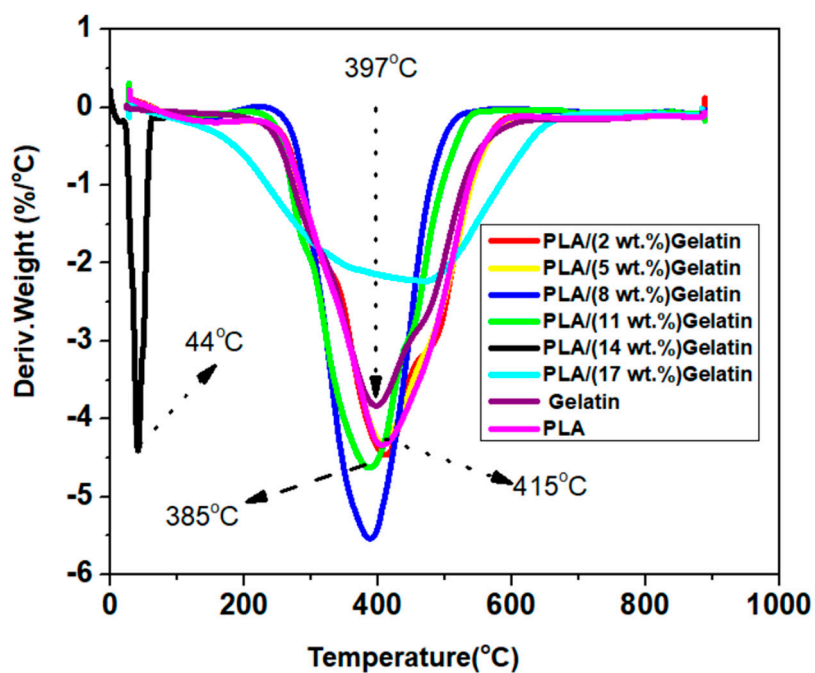


Figure 3. DTG of electrospun PLA and PLA/Gelatin Composite.

Table 1. Thermal properties of electrospun PLA, gelatin and PLA/gelatin composite.

Sample	On set	End set	Weight loss	First stage	2 nd stage	3 rd stage
PLA	302	500	70 %	-	-	-
gelatin	313	480		170 °c, & 2%	360 °c, 25%	43%
PLA/2% gelatin	321	502		203 °c, 3wt.%	377°c, 39 wt.%	502°c, 54 wt.%
PLA/5% gelatin	316	488	81wt.%	187°c, 3wt.%	-	-

PLA/8% gelatin	314	442	80WT.%	161°C, 2wt.5	-	-
PLA/11% gelatin	322	430	-	158°C, 2WT.%	361 °c, 26 wt.%	44 wt.%
PLA/14% gelatin	324	482	--	173°C, 2wt.%	357°C, 28 wt.%	50 wt.%
PLA/17% gelatin	271	533	-	-	466	38wt.%

3.3. Glass Transition Properties of Electrospun PLA and PLA/Gelatin Scaffold

The DSC of gelatin, PLA and PLA/gelatin composite is shown in Figure 4. Gelatin thermogram showed an endothermic peak. The glass transition and melting temperature were observed at 49°C and 53°C respectively. Similarly PLA glass transition and melting temperature was seen at 85°C and 172°C. which is similar to Nooeaid *et al.* (2020) report[21]. The electrospun composite exhibit more of the characteristic peak of gelatin with a T_g similar to that of gelatin, however there was a gradual shift in the T_g of the electrospun composite to higher value, with the reduction in the weight percent (8wt.%, 5wt.%, 3wt.%) of gelatin in the composite; this is assigned to increased chemical interactions resulting in the restriction in molecular movement [31]. A single T_g occurring between the T_g of two polymers is an indication of polymer miscibility. Two endothermic peak representing the melting temperature, was observed at 70 °C and 90 °C. The melting peak of gelatine undergoes significant increase in the melting temperature. The presence of two melting peak in the composite may be attributed to the formation of crystals of different sizes [32].

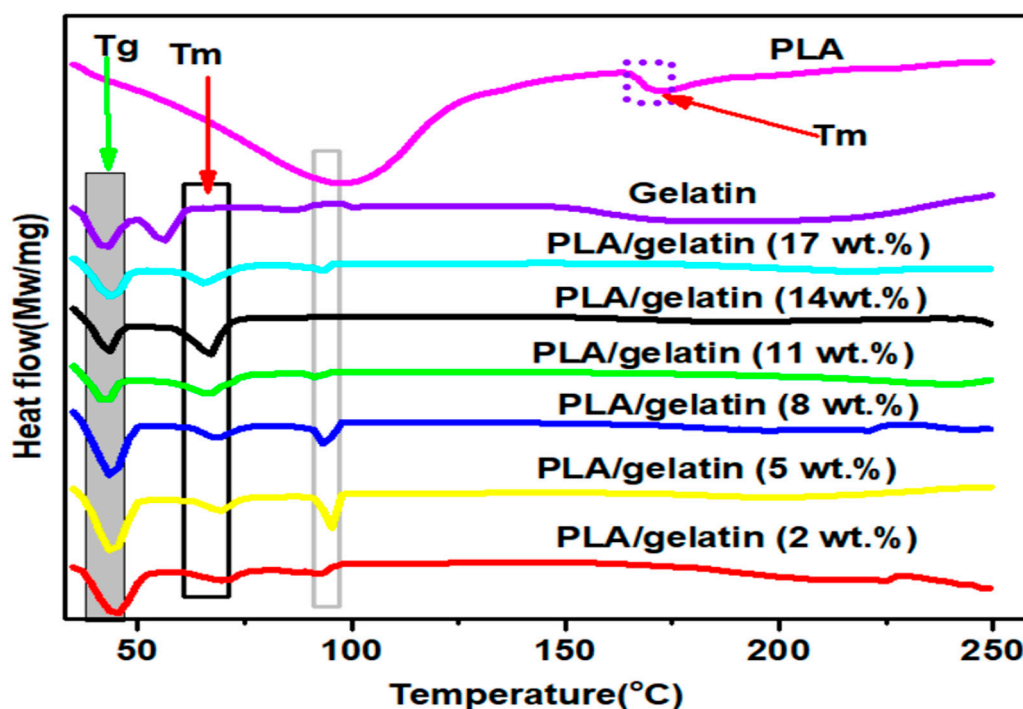


Figure 4. DSC of electrospun PLA and PLA/Gelatin Composite.

3.4. Morphological Evaluations of Electrospun PLA and PLA/Gelatin Scaffold

The morphology of electrospun PLA and gelatin is shown in Figure 5. The composite showed good fibre distribution, with enough pore spaces that allows tissue in growth and profliration. Similar morphology was also reported in the work done by Odili *et al.* (2025)[33]. The quantity of the fibres increased with increasing amount of gelatin that are present in the composite. The pore fibre diameter and pore size are shown in Figure 6. PLA/Gelatin (2 wt.%) has a fibre diameter and pore size of

6.3 μm and 0.54 pore volume fraction respectively, while PLA/Gelatin (5 wt.%) showed fibre diameter and pore size of 2 μm and 0.42 pore volume fraction respectively. increasing the amount of gelatin leads to the thinning of the scaffold fibre.

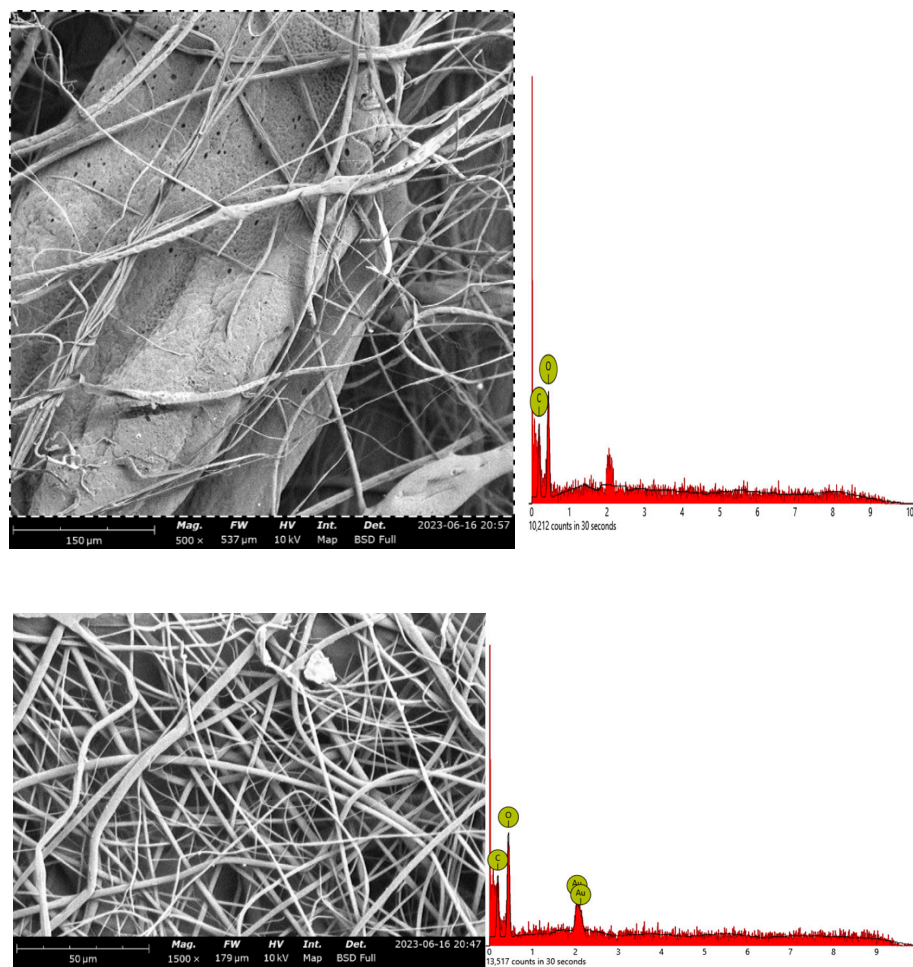


Figure 5. a: PLA/Gelatin(2 wt.%). b: PLA/Gelatin(5 wt.%). a and b: SEM morphology of electrospun PLA and PLA/Gelatin scaffold.

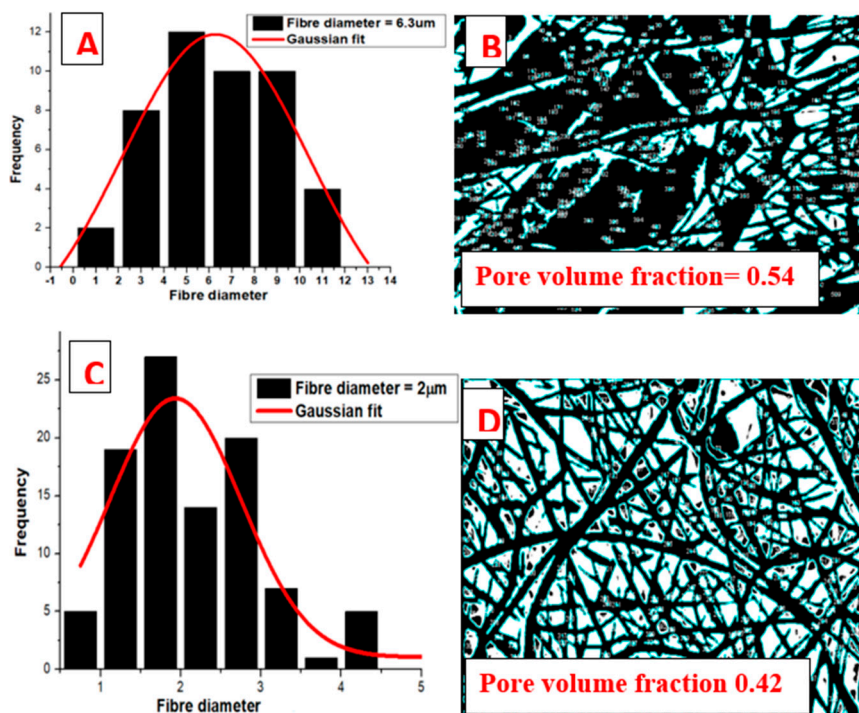


Figure 6. A and B, Fibre diameter and pore size of PLA/Gelatin (2 wt.%). C and D, Fibre diameter and pore size of PLA/Gelatin (5 wt.%).

3.5. Strength Characteristics of Electrospun PLA and PLA/Gelatin Scaffold

The tensile strength of PLA and PLA/Gelatin scaffolds are shown in Figure 7. The result reveals that the tensile strength of PLA is reduced, with increasing amount of gelatin. However, there was a spike at 14 wt.% of gelatin, thereafter the tensile strength declined. Similar observation was reported by earlier studies [8,19]. The decline can be attributed to poor bonding between the PLA and gelatin matrix. Figure 8 also showed that the elongation properties of PLA was drastically reduced with the addition of gelatin. Despite the reduction, the elongation of PLA/gelatin scaffold, improved with incremental addition of gelatin, with the highest elongation attained at 14 wt.% of gelatin.

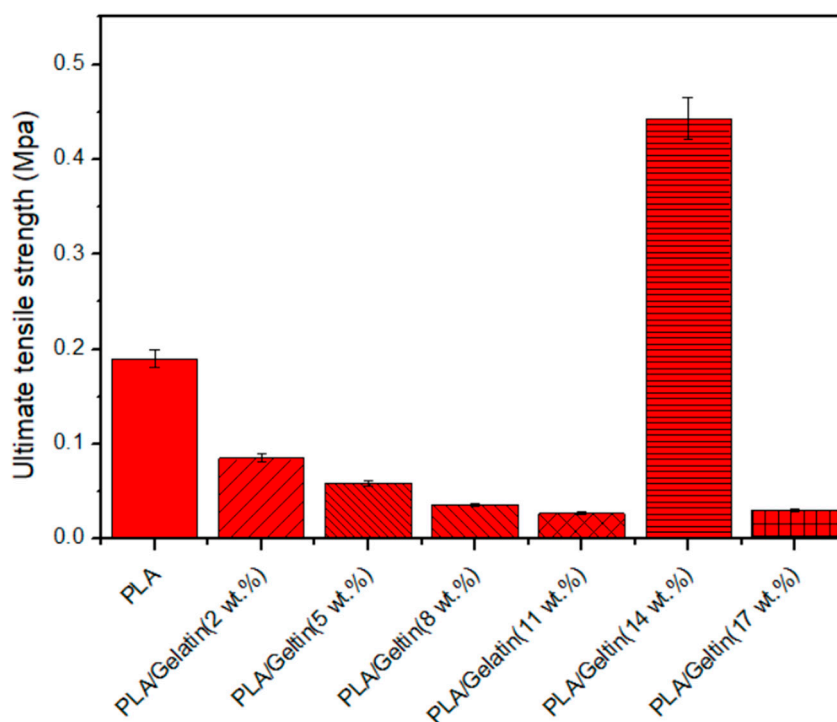


Figure 7. Ultimate tensile strength of of electrospun PLA and PLA/Gelatin scaffold.

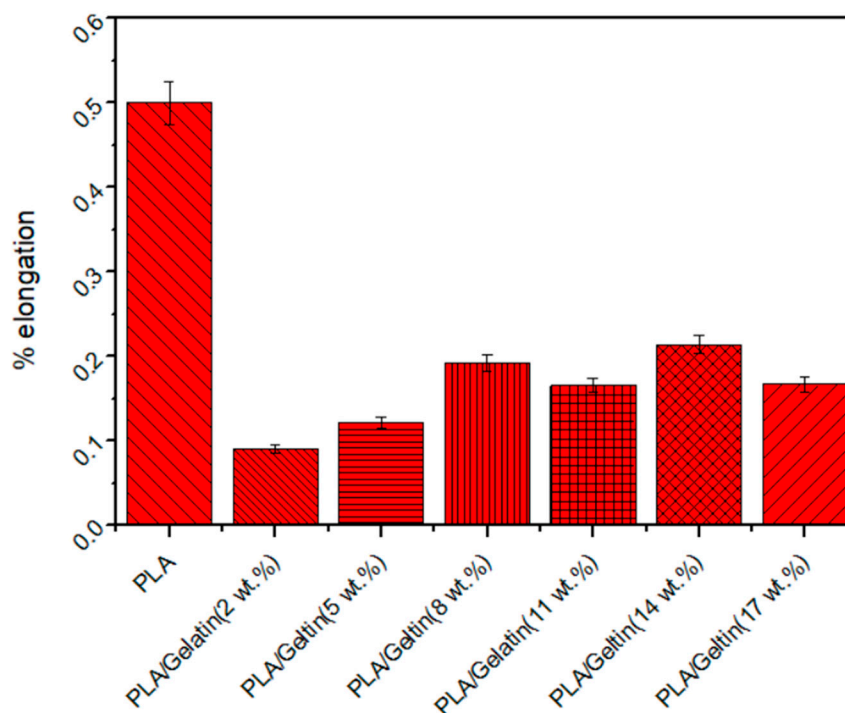


Figure 8. Percentage elongation of electrospun PLA and PLA/Gelatin scaffold.

4. Conclusions

Gelatin from fish scale was successfully extracted, incorporated into PLA and electrospun into scaffolds, forming biodegradable composites suitable for biomedical use. The interaction between PLA and gelatin was confirmed through FTIR analysis, showing characteristic peak shifts that suggest hydrogen bonding between functional groups. Thermal analysis (TGA and DSC) revealed

that the addition of gelatin enhances the thermal stability and miscibility of the composites, with peak performance observed at 14 wt.% gelatin. The SEM micrographs displayed interconnected fibrous networks with morphologies closely resembling native extracellular matrices, which are beneficial for tissue integration and nutrient transport. Although the tensile strength decreased with increasing gelatin content, the scaffold retained sufficient mechanical integrity for soft tissue application. These findings demonstrate that gelatin derived from croaker fish scale can effectively modify the physiochemical properties of PLA, providing ecofriendly and sustainable alternative to mammalian derived gelatin. The developed PLA/gelatin scaffold show promising potential for use as wound dressing materials and other tissue engineering applications, where biodegradability, hydrophilicity and biocompatibility are essential.

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Conflicts of Interest: The authors declare that there is no conflicts of interest arising from this research.:

Abbreviations

The following abbreviations are used in this manuscript:

TE	Tissue engineering
RM	Regenerative medicine

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