



## Novel Nano Biodegradable Package Films of Polycaprolactone / Extracted Gelatin from White Shavings

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*In Loving Memory of Late Professor Doctor "Mohamed Refaat Hussein Mahran"*

### Abstract

The food packaging industry is characterized by extensive reliance on synthetic plastics, which has significant implications for the environment. Consequently, there is a pressing demand for the development and implementation of biodegradable packaging alternatives. The investigation focused on examining the potential of blending polycaprolactone (PCL) and Extracted gelatin (EG) as alternative materials to non-degradable petrochemical polymers. Gelatin, as one of the most interesting natural macromolecules, is successfully prepared from protein waste. It is a biodegradable polymer commonly used in various industrial applications. In this research, gelatin resulting from leather waste production was used to form films by emulsification, producing elastic and reliable films. Gelatin was combined with polycaprolactone, a biodegradable synthesized polymer, to improve the mechanical properties of the films. The thermal behavior, physico-mechanical behavior, and topography behaviors of the samples were characterized through differential scanning calorimetry (DSC), mechanical testing, and scanning electron microscopy (SEM), respectively. In addition, Fourier transform infrared spectroscopy (FTIR) was used to examine the characteristic peaks in gelatin, PCL, and their blends. Prepared films exhibited improvements in mechanical and thermal properties, which indicated excellent applications as biodegradable packaging films. The particle size of the prepared materials was investigated through the laser scattering technique. A well-defined particle size with a narrow distribution was detected. The objective of this research is to investigate the potential of creating biodegradable films by combining extracted gelatin (EG) with a biodegradable polymer (PCL). Adding value by creating a cheap and biodegradable polymer from recycling leather protein wastes is an extreme approach to eco-friendly waste management.

Keywords: Biodegradable polymer; gelatin; packaging film; PCL

### 1. Introduction

The leather industry generates a significant quantity of waste during the hide tanning process, which is utilized to produce leather for various industries such as shoes, cars, and furniture [1-5]. The production of leather results in significant volumes of both solid and liquid waste [6, 7]. The process of manufacturing one metric ton of rawhide results in an average output of 250 kilograms of tanned leather, 250 kilograms of waste from the tanning process, 250 kilograms of untreated trash, and approximately 60,000 kilograms of wastewater [8-10]. Solid wastes produced during the leather production process include skin trimmings, keratin, fleshing, chrome shaving, and buffing wastes. These wastes primarily consist of protein, as shown in previous studies [11-13]. The issue has garnered increased attention due to its predominant disposal in landfills up until the present moment. Failure to utilize, process, or adequately manage protein-based solid waste can give rise to significant ecological

challenges that pose a threat to the long-term viability of the leather industry. An increasing number of effective methods for treating solid and liquid waste have been shown to have important benefits for both the economy and the environment [14-16]. The primary waste products of the leather industry, known as white leather shaving fibers (WLSF), are processed into several valuable compounds, including glue and gelatin, for industrial use [11]. The potential of incorporating gelatin into polymeric materials was investigated, with promising outcomes. There is a growing interest in the extraction of gelatin and its derivatives, driven by the increasing trend of substituting synthetic agents with gelatin in various industrial processes. This shift towards gelatin use contributes to the enhanced utilization of by-products generated from animal slaughter. The properties of gelatin are contingent upon the specific raw material utilized and the conditions under which it is extracted. These factors ultimately dictate the range of applications for which gelatin can be employed.

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Gelatin is utilized in a diverse range of industries, including the culinary, pharmaceutical, cosmetic, and packaging sectors [17, 18].

In contemporary packaging technology, synthetic polymers pose numerous challenges due to their limited capacity for recycling [19]. The proper management of a significant quantity of packaging waste necessitates the utilization of high temperatures during landfill disposal or incineration processes. This is crucial in order to prevent the generation of degradation byproducts that possess undesirable characteristics, often including carcinogenic traits. The presence of such byproducts can have a negative impact on the overall expenses associated with waste disposal. Synthetic polymers have been observed to undergo a prolonged breakdown process lasting several centuries when disposed in landfills [20-23]. Polyamide (PA), polypropylene (PP), polyethylene (PE), and polystyrene (PS) are commonly employed petroleum-based polymers for packaging applications. Plastic has achieved significant success in the packaging industry due to its exceptional mix of strength, transparency, stability, and flexibility. These qualities render it extremely ideal for addressing persistent environmental concerns, particularly in the context of food packaging [20].

On the contrary, while possessing favorable attributes, the utilization, gathering, and consolidation of those assets entail significant environmental hazards, due to their reliance on fossil fuels [24]. The current global consumption of plastics is rapidly reaching 100 million metric tons per year, which has now emerged as a pressing issue of great importance. In order to identify a substitute for plastic, it is imperative to explore options derived from renewable and recyclable sources. In response to the escalating issue of plastic waste and pollution, researchers have directed their efforts towards enhancing substitute packaging materials derived from renewable resources that possess biodegradable or compostable properties [25]. Various types of biodegradable plastics and packaging materials have already been developed, including cellulose, starch, and protein-based alternatives. Biodegradable polymers, characterized by their ability to undergo decomposition through natural processes, are the focus of this discussion. The degradation period of viruses, clamps, or weather in landfills, typically measured in weeks or months, presents a highly appealing alternative within this domain. Furthermore, these polymers are derived from naturally occurring sources, specifically renewable raw resources such as animal or plant proteins, and possess non-toxic properties [26]. Biopolymers are employed as substitutes for non-biodegradable plastics, thereby introducing a novel category of materials known as bioplastics. This transition to bioplastics serves to mitigate environmental impact and reduce reliance on oil resources [27]. Bioplastics can be categorized into three primary groups based on

their source and biodegradability. The first group consists of biobased non-biodegradable bioplastics, such as PET and PA. The second group includes biobased biodegradable bioplastics like PLA, polyhydroxyalkanoates (PHA), starch, and gelatins [28]. The third group comprises fossil-based biodegradable bioplastics, including polycaprolactone (PCL) [29, 30]. According to European Bioplastics, biobased materials can be classified into two categories: those derived from biomass, such as PS and gelatin, and those derived from petrochemicals, such as PLA and PCL [31]. In the context of dietary requirements, it is crucial to adjust the mechanical properties of biopolymer-based materials in order to enhance their functional capabilities. These adjustments may have been achieved through crosslinking, which involves physical or chemical alterations, as well as blending with additional components such as plasticizers or compatibilizers [32]. Gelatin exhibits an amorphous structure [33]. Polycaprolactone (PCL) is a biodegradable polyester with a semicrystalline structure [34]. The quality of films is contingent upon both the origin and the handling of the material. Additionally, the surrounding conditions have a substantial role in influencing the mechanical properties of the material.

Gelatin is the main extracted target in protein shaving acid or alkaline hydrolysis process [35]. The mechanical properties of extract gelatin are weak characterized as a film. For that, many research groups are focus on blending or grafting for different polymers with gelation to increase its mechanical properties [36, 37]. Due to biomolecular behavior of gelatin combing with biodegradable polymers especially polylactic acid, polyglycolic acid and polycaprolactone is one of hot topics for various applications [38, 39].  $\epsilon$ -caprolactone (CL) is a commonly known as cheap monomer [40]. Polycaprolactone (PCL) was prepared through ring-opening polymerization of cyclic monomer in presence of Tin(II) 2-ethylhexanoate catalyst [41-44]. PCL can easily biodegradable through action of fungi and enzymes[45]. The rate of degradation can be controlled through copolymerization of PCL with glycoside or lactide [46]. Blending of gelatin with PCL and polyethylene glycol as a biodegradable hydrogel scaffolds has been studied [47]. Extensive study of the effect of a solvent on the compatibility of PCL/gelatin was performed. FTIR and visco-mechanical measurements exhibited that the kind of solvent effects the structure of PCL/gelatin polymer molecules in solution. PCL/gelatin films and membranes were prepared through electrospun and/or casting [48]. In addition, Guarino et al. [49] applied the combination of PCL and gelatin by contact layer over layer by solvent cast of gelatins and electrospun of PCL. The barrier coating, topography, antimicrobial, mechanical, thermal, water vapor and oxygen permeability properties were studied. Increasing

homogeneity of PCL and gelatin blend can be accomplished by adding of gelatin–polycaprolactone blend. The prepared polymer blends exhibited enhanced flow and thermal behavior in the variety between the different polymer ratios [50, 51].

This article focused on one of the major environmental waste problems of the huge manufacture in Egypt and over the world (leather industry). The objective of this research is to investigate the potential of creating biodegradable films by combining extracted gelatin (EG) with a biodegradable polymer (PCL). Large amounts of solid wastes containing proteins are the main wastes resulted from lather tanning process. We extracted high value gelatin from white leather shavings. In addition, preparation of biodegradable packaging films as a new challenge is a plastic film production. Moreover, this project focused on using extracted gelatin to produce biodegradable packaging films in presence of selected synthesized biopolymers. Overall, this research idea studied vital environmental and industrial problems and share in new research fields by preparing eco-friendly packaging materials.

## 2. Experimental

### 2.1 Materials and methods

Tin(II) 2-ethylhexanoate ( $\text{Sn}(\text{Oct})_2$ ), triphenylphosphine and glycerol was purchased from Sigma-Aldrich, München, Germany.  $\epsilon$ -caprolactone (Sigma-Aldrich, München, Germany) was dried and purified before use by storage over  $\text{CaH}_2$  for 24 h, followed by distillation under low vacuum. Dichloromethane (DCM), chloroform and methanol (Sigma Aldrich, Missouri, USA) are used after drying over  $\text{CaH}_2$ . Extracted gelatin from white leather shavings was supplied and discussed by us [11].

### Methods

#### Differential scanning calorimetry (DSC)

Differential scanning calorimetry DSC131 evo (SETARAM Inc., France) was used to perform the differential scanning calorimeter analysis, Nanomaterial Investigation laboratory, Central Laboratories Network, National Research Centre (NRC), Egypt. The instrument was calibrated using the standards (Mercury, Indium, Tin, Lead, Zinc and Aluminum). Nitrogen and Helium were used as the purging gases. The test was programed including the heating zone from  $-50^\circ\text{C}$  to  $200^\circ\text{C}$  with a heating rate  $10^\circ\text{C}/\text{min}$ . The samples were weighted in Aluminum crucible 100  $\mu\text{l}$  and introduced to the DSC. The thermogram results were processed using (CALISTO Data processing software v.149).

### Nanoparticle characterizations

The mean diameters investigation of dispersed samples was carried out with dynamic laser scattering (DLS) instrument (PSS, Nicomp, USA), using the 632 nm HeNe laser with incident angle of light  $170^\circ$ .

### Synthesis of gelatin nanoparticles

The double desolvation technique was used for preparation of gelatin nanoparticles [17] with our essential adaptations. 6.1 wt.% gelatin solution was prepared by dissolving 3.05 g of gelatin in 50 mL Milli-Q water with warming condition with magnetic stirring. The desolvation step was carried out by adding 60 mL of acetone to the gelatin solution. Followed by re-desolvation in Milli-Q water and lyophilizing the precipitated gelatin fraction. A 2 % gelatin solution was obtained via dissolving 0.2 g of the lyophilized gelatin in 20 mL of Milli-Q water at  $50^\circ\text{C}$ . The dropwise addition of acetone under continuous stirring 800 rpm for 12 h was leading to formation of nanoparticles. The purified nanoparticles were prepared through three-steps centrifugation 40,000 rpm for 15 min, and redispersed in acetone/water (30/70 mixture). The resulted dispersion of nanoparticles in Milli-Q water was kept in refrigerator.

### Preparation of biodegradable packaging film

PCL solution was prepared by dissolve 25 mg of polymer were dissolved in 5 mL of chloroform. Polymer solution was added to 10 ml of Millipore water drop wise. The mixture was stirred overnight to allow remove of chloroform. Resultant solution containing PCL nanoparticles were blended with different weight ratios, 1:1, 1:2 and 2:1 of extracted gelatin in presence of 2 wt% glycerol used as plasticizer and 0.15 wt% triphenylphosphine as edible emulsifier. The mixture stirred for 60 min followed by film casting in Teflon petri-dish and dried in vacuum oven at  $30^\circ\text{C}$  overnight to use as biodegradable packaging films.

### ATR-FTIR investigation

The structure change between gelatin and gelatin/PCL was studied by attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectroscopy (PerkinElmer, Inc., Shelton, USA). All spectra were measured in wave numbers ranging from  $450$  to  $4000\text{ cm}^{-1}$  [52].

### Scanning Electron Microscopy (SEM)

Field emission scanning electron microscopy (Quanta 250 FEG, FESEM) was carried out for PCL and PCL/gelatin samples. The specimens were processed with Au spraying before imaging [53].

### The mechanical properties

The mechanical properties (tensile strength and elongation % at break) were determined to ASTM D638-10, Type I (ASTM, 2010). All the studied dry films were cut into strips. Five strip samples of each film type were desiccated at 33% RH for 24 hours at ambient temperature. The strip samples were measured on an Instron Universal Testing Machine at a crosshead speed of 15 mm/min and 0.1 kN force load

cell.

## Results and Discussion:

### ATR-FTIR analysis

The Fourier Transform Infrared (FTIR) spectra exhibited distinctive absorbance peaks corresponding to the chemical groups present in gelatin, polycaprolactone, and various blends of polycaprolactone with gelatin at varying ratios (1/1, 1/2, 2/1) in Figure 1 and Table 1.

The identification of absorbance peaks is often conducted in the following manner: The presence of a broad and intense band in the range of 3600-3200  $\text{cm}^{-1}$  can be attributed to the peptide bond (NH) and the hydroxyl group (OH) of the carboxylic group in gelatin [54, 55]. Additionally, the bands observed at 2952  $\text{cm}^{-1}$  and 2848  $\text{cm}^{-1}$  can be attributed to the stretching vibrations of -CH<sub>3</sub> and -CH<sub>2</sub> groups in both gelatin and polycaprolactone. The strong bands observed at 1660  $\text{cm}^{-1}$  and 1720  $\text{cm}^{-1}$  are indicative of the overlapping stretching vibrations of the carbonyl groups in gelatin and polycaprolactone. Furthermore, the bands observed at 1388  $\text{cm}^{-1}$  and 1434  $\text{cm}^{-1}$  can be attributed to the bending vibrations of -CH<sub>3</sub> and -CH<sub>2</sub> groups [56].

The band associated with the NH group was observed at a wavenumber of 1237  $\text{cm}^{-1}$ , indicating the presence of amide functional groups. Additionally, a prominent band at 1160  $\text{cm}^{-1}$  was observed, which can be related to stretching vibrations of the -C-O-C- bonds in polycaprolactone. The bands observed at wavenumbers of 987  $\text{cm}^{-1}$ , 969  $\text{cm}^{-1}$ , and 695  $\text{cm}^{-1}$  exhibit a correlation with the presence of the -C-C- functional group, as depicted in Figure 2.

In conclusion, the presence of distinct and extensive overlapping bands within the spectral range of 3200-3600  $\text{cm}^{-1}$  in the polycaprolactone/gelatin blended ratios of (1-1, 1-2, 2-1) provides compelling evidence of a highly effective chemical blending of the composite materials. Furthermore, the presence of distinct and well-defined bands at 1660  $\text{cm}^{-1}$  and 1720  $\text{cm}^{-1}$ , which can be attributed to the C=O stretching vibration of gelatin and polycaprolactone, provides evidence for the successful blending of these materials.

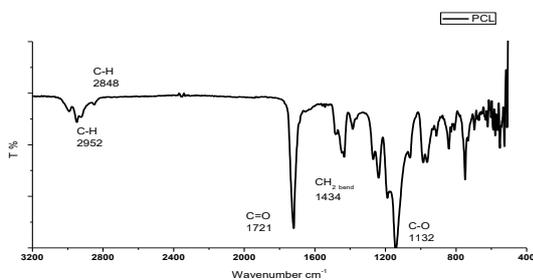


Figure 1a. FTIR spectrum of PCL as parent in

### biodegradable packaging film

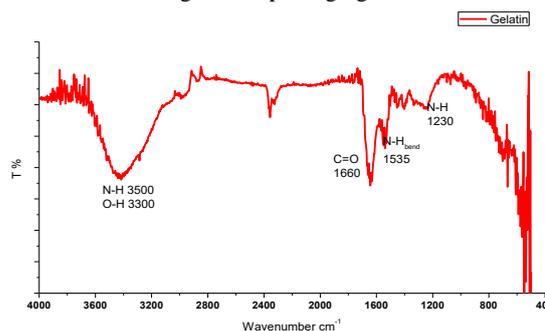


Figure 1b. FTIR spectrum of extracted gelatin as parent in biodegradable packaging film

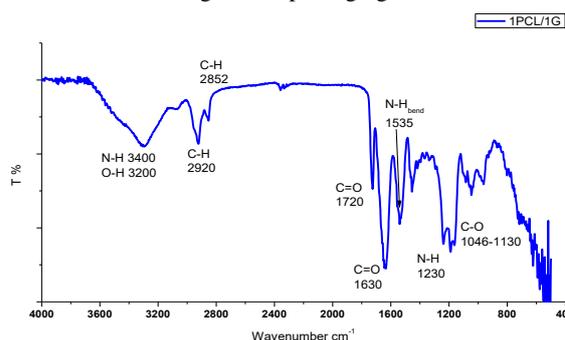


Figure 1c. FTIR spectrum of gelatin/PCL with ratio 1:1 in biodegradable packaging film

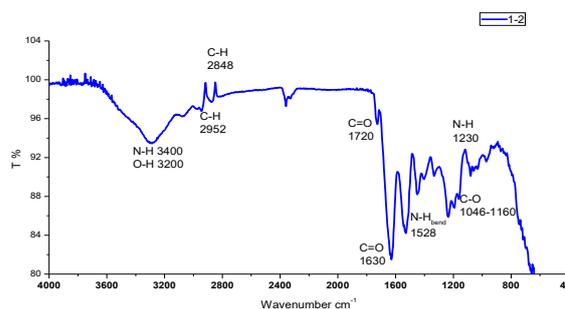


Figure 1d. FTIR spectrum of gelatin/PCL with ratio 1:2 in biodegradable packaging film

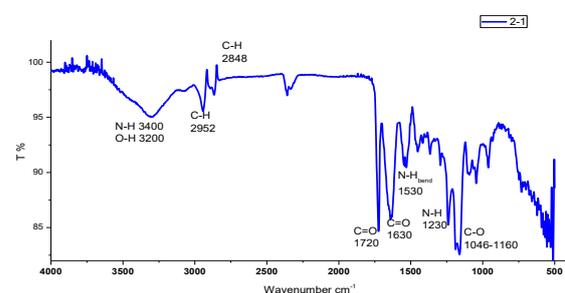


Figure 1e. FTIR spectrum of gelatin/PCL with ratio 2:1 in biodegradable packaging film

Table 1: FTIR spectra for polycaprolactone, technical gelatin and the blended ratio (1/1, 1/2, 2/1).

Assignment	Peak wave number $\text{cm}^{-1}$		
	Gelatin	Polycaprolactone	Polycaprolactone blended with gelatin
NH stretch	3500	-----	3500
OH stretch of water, gelatin	3340	-----	3346
CH <sub>2</sub> asymmetrical stretch	2930	2952	2930
CH <sub>2</sub> symmetrical stretch	2850	2848	2852
C=O stretch, coupled with COO- gelatin	1660	1721	1720, 1660
NH bend coupled C-N stretch	1535	-----	1535
CH <sub>2</sub> bend	1450	1451	1452
CH <sub>2</sub> wagging of proline	1338	-----	1335
NH bend	1230	-----	1235
C-O stretch	1046	1160	1040-1160
C-C stretch		1000	
CH out of plane, skeletal stretch	593	959, 841, 748	861, 674, 618

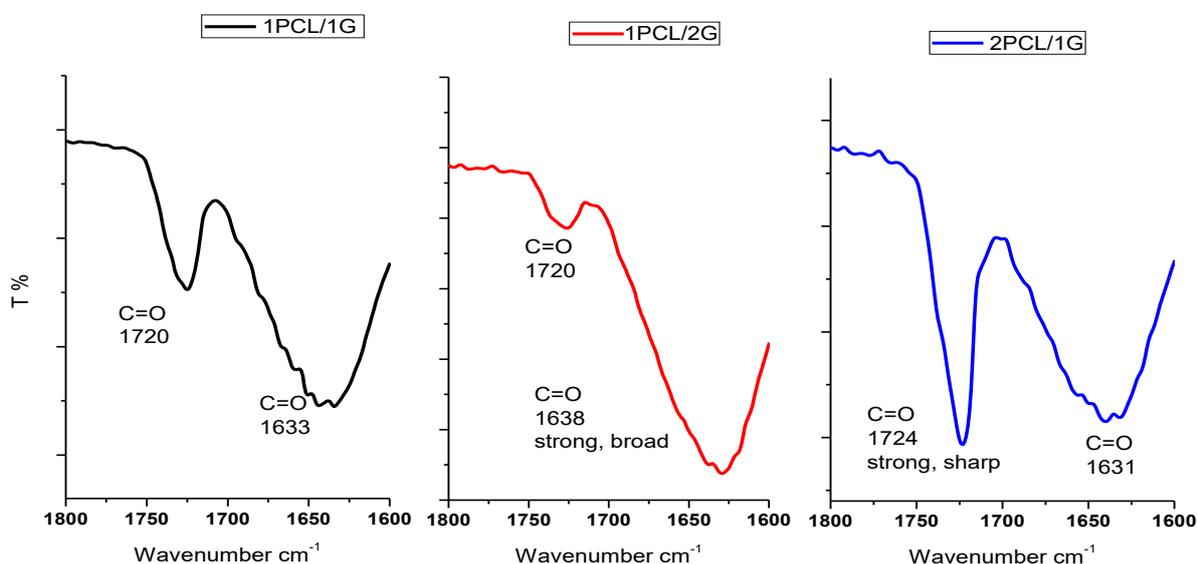


Figure 2. Amide I and Amide II band for Gelatin / PCL blended with different ratios

### Solvent Casting Film

Solvent casting is often regarded as the predominant process for film formation, particularly in large-scale industrial applications [56]. The production of film casting can involve the creation of both thick and thin films by manipulating the solid content of the solution. Furthermore, the choice of solvent significantly influences the texture of the film and the level of uniformity achieved when combining polymers [57]. The mechanical and morphological qualities of the casted film can be improved by optimizing the casting temperature and drying conditions. The solvent casting process was employed to fabricate poly( $\epsilon$ -caprolactone) (PCL) films, and a coater with top heating cover (MSK-AFA-III, China) was utilized to blend PCL with gelatin films.

### Morphological structure

The morphological characteristics of PCL were examined using scanning electron microscopy (SEM)

both before and after its combination with gelatin recovered from protein wastes. The SEM images are presented in Figure 3. The analysis revealed that the topographic images indicated that PCL exhibited a uniform shape. The roughness of the film surface exhibited an upward trend as the gelation percentage in PCL/gel films increased. This finding suggests that the interaction between gelatin and PCL polymer was predominantly homogenized.

The examination of the surfaces' topography and texture of the specimens was conducted using a SEM, as depicted in Figure 3. The PCL/gelatin mixtures, namely in ratios of 1:1, 1:2, and 1:3, are prepared by combining the components according to the weight of the extracted gelatin. The surface of the raw PCL, as depicted in Figure 3a, had a clean and relatively smooth appearance, with the exception of a few holes. These holes mimicked the characteristic brittle fracture mechanism observed during solvent evaporation.

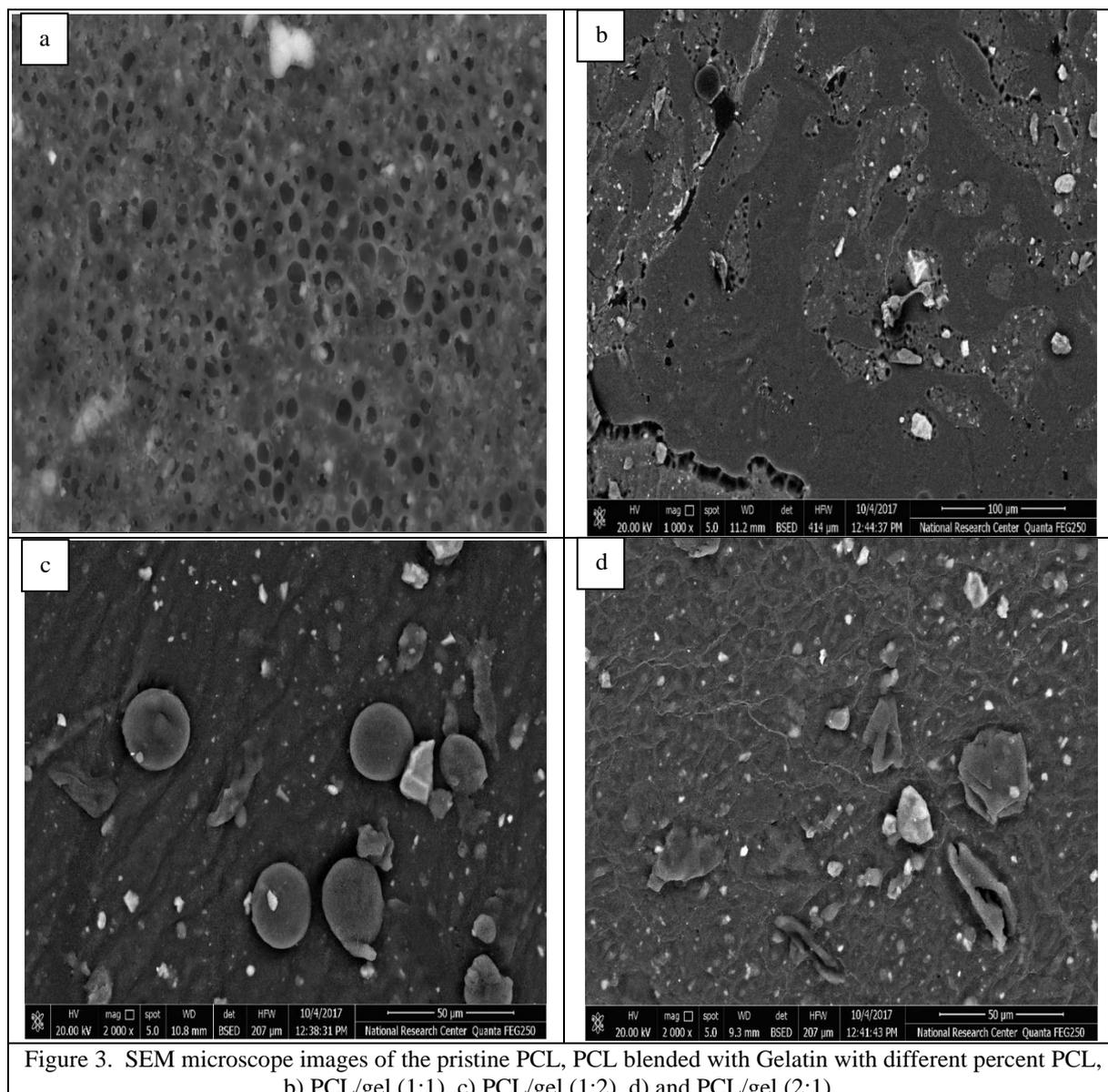


Figure 3. SEM microscope images of the pristine PCL, PCL blended with Gelatin with different percent PCL, b) PCL/gel (1:1), c) PCL/gel (1:2), d) and PCL/gel (2:1) .

The roughness of the fracture surface of the PCL/gel was shown to be greater compared to that of the raw PCL, as depicted in Figure 3b. The specimens exhibited small surface heterogeneities resulting from the solvent casting procedure. Additionally, they displayed a dense bulk phase that was initially well-compacted but subsequently lost its organization during solvent evaporation, as depicted by the arrow in Figure 3b. Multiple coarse fracture surfaces and a greater quantity of dispersed phases were observed in the gelatin samples (Figures 3c to 3d). As the concentration of gelatin within the PCL matrix grew, the dispersion of gelatin became increasingly uniform. Furthermore, the formation of a continuous phase consisting of polycaprolactone (PCL) and gelatin was achieved in a biodegradable film composed of PCL and gelatin. Nevertheless, it was discovered that when the mass proportion of gelatin exceeded 50% in the combined PCL/gel system, the formation of gelatin

aggregates occurred, resulting in phase separation and ultimately leading to a brittle nature.

#### Differential Scanning Calorimetry (DSC)

DSC measurements proved to be a valuable technique for assessing the blending efficiency in multi-component systems. The glass transition temperatures ( $T_g$ s) of each individual component were seen to exhibit a desirable level of uniformity. This was achieved by reducing the spatial separation between the  $T_g$ s of two or more components.

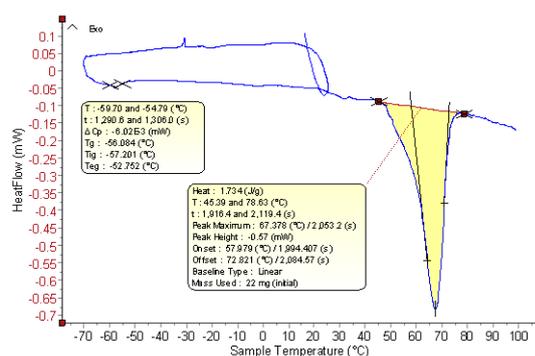


Figure 4a. Differential scanning calorimetry (DSC) thermograms of PCL

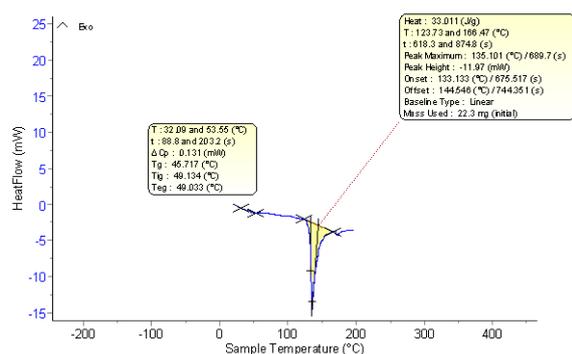


Figure 4b. Differential scanning calorimetry (DSC) thermograms of gelatin

Polycaprolactone (PCL) exhibits crystalline and/or semi-crystalline characteristics depending on the synthesis methods employed and the precipitation process. Differential scanning calorimetry (DSC) was employed to assess the level of miscibility shown by the scaffolds. The melting point of PCL is observed to occur at a temperature of 60 °C, whereas its glass transition temperature ( $T_g$ ) is measured to be -60 °C. Nevertheless, the gelatin that was extracted exhibited degradation at a temperature of approximately 137 °C, which is close to its melting point. In Figure 4a, it is observed that the glass transition temperature ( $T_g$ ) of the PCL sample developed by our research team is measured to be -56 °C, whereas the melting point is determined to be 57 °C. The obtained results indicate that the glass transition temperature ( $T_g$ ) of the extracted gelatin gel was measured to be 45 °C, whereas the melting point was determined to be 133 °C, as seen in Figure 4b. Figure 5 demonstrates that the melting temperature ( $T_m$ ) of PCL decreases as the percentage of gelatin increases in PCL/gelatin blends. The reduction of the melting temperature ( $T_m$ ) can be attributed to the chemical and/or physical interaction occurring between gelatin and PCL in the blend.

The poly( $\epsilon$ -caprolactone) (PCL) exhibited a phase transition at a temperature of -56 °C. This transition was ascribed to the glass transition temperature ( $T_g$ ) as well as a melting process occurring within the temperature range of 40 °C to 66 °C, with a melting temperature ( $T_m$ ) of 57.7 °C. The melting process was

shown to be associated with the formation of a crystalline phase. In the context of gelatin blending, the observed rise in gelatin percentages with a glass transition temperature ( $T_g$ ) almost equal to 45 °C indicates a corresponding decrease in the  $T_g$  of the PCL at lower temperatures, as depicted in Figure 5. The increase in gelatin content resulted in a corresponding increase in the glass transition temperature ( $T_g$ ) of the blend. Specifically, the  $T_g$  increased from -60 °C for pure poly( $\epsilon$ -caprolactone) (PCL) to 22 °C for the PCL/gelatin blend in a 1:1 ratio, as depicted in Figure 5 and Table 2.

Table 2: Differential scanning calorimetry (DSC) of PCL, PCL blended with Gelatin with different percent PCL, PCL/gel (1:1), PCL/gel (1:2), and PCL/gel (2:1).

Polymer	(DSC) thermograms $T_g$ °C
PCL	-60
PCL/gelatin*(2:1)	-45
PCL/gelatin*(1:2)	-35
PCL/gelatin*(1:1)	22

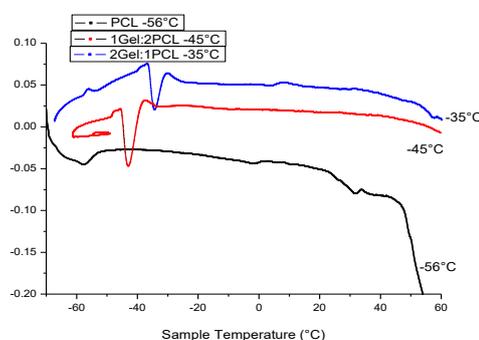


Figure 5. Differential scanning calorimetry (DSC) of PCL, PCL blended with Gelatin with different percent PCL, PCL/gel (1:2), d) and PCL/gel (2:1).

In conclusion, differential scanning calorimetry (DSC) analysis provides a reliable indication and validation of the successful blending of two components, particularly polycaprolactone (PCL) and gelatin, demonstrating their complete miscibility. A newly identified glass transition temperature ( $T_g$ ) was observed within the range including the  $T_g$  values of the individual constituent materials in the blend.

### Dynamic light Scattering (DLS)

Differential light scattering (DLS) exhibits a high level of detection effectiveness in assessing both the particle size distribution and the degree of variety. The particle size distribution of dispersed PCL, gelatin, and PCL/gel in an aqueous medium exhibits a limited polydispersity, as depicted in Figure 6. The measured particle size ranges from 0.45 to 1.6  $\mu\text{m}$ , with over 90% of particles by volume falling within this size range, as indicated in Table 3.

The correlation between the uniform dispersion of blended films gelatin and the reduced variability in particle size and polydispersity of PCL is obvious.

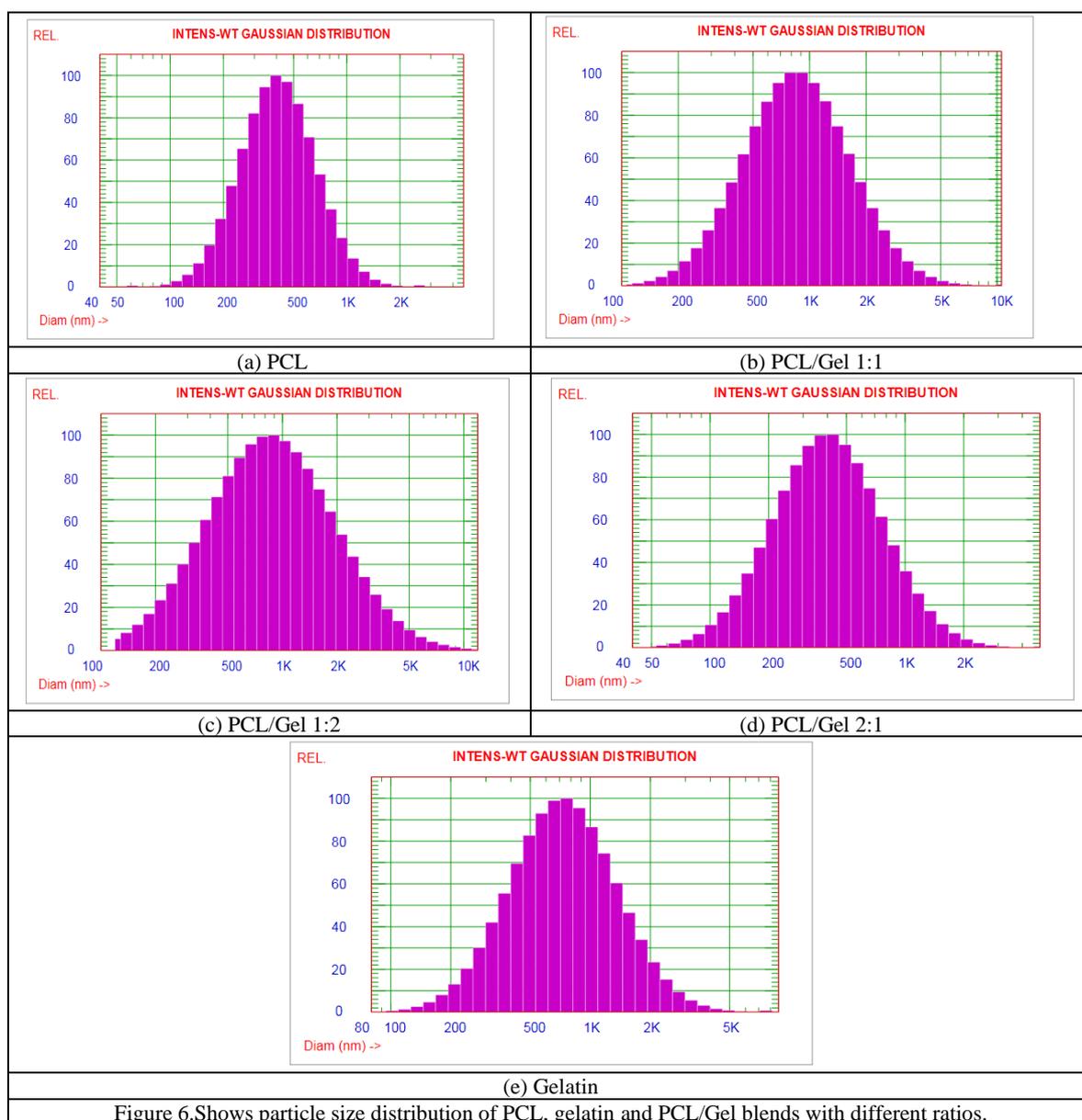


Figure 6. Shows particle size distribution of PCL, gelatin and PCL/Gel blends with different ratios.

Table 3. Particle size distribution and mean particle size of PCL, Gelatin and combines with different ratios

Sample	Mean diameter, nm	Variance(PI)	Standard deviation	Chi Square
PCL	492.0			
PCL/Gel 1:1	1056.2	0.432	693.3 nm (65.7%)	91.233
PCL/Gel 2:1	458.8	0.242	225.7 nm (49.2%)	8.357
PCL/Gel 1:2	1193.9	0.667	907.4 nm (81.7%)	98.014
Gel	879.5	0.382	543.5 nm (61.8%)	4.791

### Mechanical Properties

In the succeeding phase, Poly( $\epsilon$ -caprolactone) (PCL), gelatin, and PCL/gelatin blends with varying ratios were integrated into a PCL matrix to fabricate blended films using the solvent casting method. The most significant enhancement was noticed in the case of PCL/gelatin film with a weight ratio of 2:1 and 1:2. The results shown in Table 4 demonstrate that the premature modulus and mechanical tensile properties of the PCL film were enhanced by 223.3% and 159%, respectively, when compared to the raw PCL film. The

obtained results indicate a consistent film thickness of approximately  $\pm 43 \mu\text{m}$  for PCL films and  $\pm 1.5 \mu\text{m}$  for PCL/gel films. The behavior of particle size enables the application of PCL and PCL/gel film in a wide range of areas, including packaging films with sizes as small as a few  $\mu\text{m}$ . The observed enhancements can be attributed to the increased interaction and flexibility between the gelatin and PCL matrix, achieved through an increase in the sliding coefficient between the polymer chains.

Table 4. Physico-mechanical properties of PCL, gelatin and PCL/gel blends with different ratios.

Sample	Stress, MPa	Tensile, MPa	Elongation, %	Elasticity
PCL	7.4	9.4	0.9	brittle
PCL/gel 1:1	12.33	15.59	297	elastic
PCL/gel 2:1	14.72	19.34	213	Semi elastic
PCL/gel 1:2	9.65	13.15	332	High Elastic
gel	8.44	11.28	190	Elastic

The blended sample with a PCL to gelatin ratio of 2:1 exhibits the highest mechanical characteristics. The high loading force applied during mechanical measurement has a significant impact on the high flexibility of gelatin chains above a certain percentage. The rapid expansion of blended film does not allow for significant elongation prior to sample cutting. The observed correlation between the applied force and the measured tensile strength and elongation at break of the blended sample suggests potential applications for the created polymer film as biodegradable packaging films.

### Conclusion

The leather processing companies produces substantial quantities of fibrous waste, which can be economically reused through the chemical degradation recovery of WLSF. The gelatin isolated from a substance was combined with the PCL matrices. Blends including gelatin and polycaprolactone (PCL) were homogenized and created by the process of casting film creation. The intermediate low molecular weight of EG allowed for effective mixing with PCL, as observed through SEM analysis. The morphology of films was characterized to determine the presence of interphase formation. This was accomplished by evaluating the particle diameter as a function of the increasing amount of PCL. The elongation at break percentage of PCL/EG thermoplastic films increased as the percentage of EG rose to 66 wt %. The thermoplastic films exhibited excellent elasticity, with an elongation at break of 332%. However, the stress showed no significant influence. These properties make the films potentially suitable for applications in the packaging sector. The films that were created demonstrated notable thermal stability when the PCL ratios in the blends were increased. Additionally, the dynamic light scattering (DLS) analysis demonstrated that there was a restricted impact on both the average particle size and the polydispersity characteristics of the polycaprolactone (PCL) material, thereby verifying its compatible behavior. The mechanical properties of the cast films were found to fall within an acceptable range for packaging film. The use of polycaprolactone (PCL) enhances the topographic properties of biodegradable composite films, so making them more suitable for use as environmentally-friendly packaging materials. Through an examination of the thermal, mechanical, and morphological characteristics of the manufactured biodegradable films PCL/EG, it was determined that

the inclusion of 50 wt % of EG resulted in blended films that exhibited favorable thermal processability, transparency, flexibility, and mechanical properties. The thermal degradation of PCL was influenced by the incorporation of gelatin ranging from 30 to 66 weight percent. It was determined that the use of gelatin derived from the leather industry for the creation of biodegradable films was feasible and promising. The positive results obtained enable the creation of low-cost sustainable packaging films.

### Competing interests.

The authors have declared no conflict of interest.

### Compliance with Ethics Requirements

This article does not contain any studies with human or animal subjects.

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### Research Ethics

We are not required to complete an ethical assessment prior to conducting your research. Animal ethics: we are not required to complete an ethical assessment prior to conducting your research.

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