



Research Paper / Makale

Preparation and Characterization of Dialdehyde Cellulose/Polylactic Acid Blends

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Abstract: In recent years, the demand for naturally derived synthetic materials to protect the environment has increased. Cellulosic polymers are renewable, biodegradable, eco-friendly, and possess high strength and stiffness. As a result of the researches, the use of polylactic acid (PLA) instead of petroleum-based polymers stands out as an alternative. In this study, dialdehyde cellulose (DAC)/polylactic acid (PLA) mixtures were prepared. DAC was prepared by peroxide oxidation from microcrystalline cellulose. Firstly, DAC was blended with PLA. In addition to having hydrophilic properties of DAC, polyethylene glycol (PEG) was added to increase compatibility between PLA and DAC, as PLA was known to have hydrophobic properties. The blends were characterized by infrared spectroscopy (FT-IR) and thermal analysis (TGA and DTG) techniques. Besides, scanning electron microscopy (SEM) images of the polymer blends obtained were interpreted.

Keywords: Biodegradable polymers, blend polymers, cellulose, dialdehyde cellulose, polylactic acid, termal analysis

Dialdehit Selüloz/Polilaktik Asit Blendlerinin Hazırlanması ve Karakterizasyonu

Öz: Son yıllarda çevreyi korumak için doğal olarak türetilmiş sentetik malzemelere olan talep artmıştır. Selülozik polimerler doğada yenilenebilir, biyolojik olarak parçalanabilir, çevre dostu olmalarının yanında yüksek mukavemet ve sertliğe sahiptirler. Yapılan araştırmalar neticesinde, petrol bazlı polimerler yerine polilaktik asit (PLA) kullanımı alternatif olarak öne çıkmaktadır. Bu çalışmada biyobozunur polimer olan selülozdan yola çıkılarak dialdehit selüloz (DAC)/polilaktik asit (PLA) karışımları hazırlandı. DAC, mikrokristalin selülozdan peroksit oksidasyonu ile elde edildi. İlk olarak DAC/PLA blendi hazırlandı. DAC hidrofilik özelliklere sahip olmasının yanında, PLA'nın hidrofobik özelliklere sahip olduğu bilindiğinden, PLA ve DAC arasındaki uyumluluğu artırmak için polietilen glikol (PEG) eklendi. Blendler, FT-IR spektroskopi ve termal analiz (TGA ve DTG) teknikleriyle karakterize edildi. Ayrıca elde edilen polimer karışımlarının SEM görüntüleri de yorumlandı.

Anahtar Kelimeler: Biyobozunur polimerler, blend polimerler, dialdehit selüloz, polilaktik asit, selüloz, termal analiz

1. Introduction

Polymer materials are indispensable elements in our daily life. Environmental pollution, the high price of crude oil, and global warming have increased interest in replacing petroleum-based polymers with biodegradable biopolymers. Moreover, much effort has been made to develop advanced materials for solving these problems [1].

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Biodegradable polymers were first introduced in the 1980s. Polymers from renewable sources have gained increasing interest in the past two decades, mainly for two main reasons: first, their environmentally hazardous effects, and second, our oil resources are limited [1-3]. Biodegradable material; it is possible to identify it as a biodegradable substance. Biodegradability is defined by the American Society for Testing and Materials (ASTM) as follows; "Materials are predominantly determined by standard tests (ASTM 2005) which can decompose to biomass, inorganic components, water, methane, carbon dioxide by enzymatic reactions of microorganisms and reflect their decomposition at a certain time". Biodegradable polymers can be produced either naturally or from the biological system and are non-toxic. They are used very frequently, especially in the field of food and medicine [4].

Considering the existing polymers synthesized, scientists modify these materials' structures in different ways to suit their wishes. One way of obtaining these materials is by polymer blending. The polymer blend can be defined as a homogeneous or heterogeneous mixture of at least two polymers/copolymers. These polymers can interact chemically or physically, taking into account their structural properties, and their physical properties are different from their main components [5].

Cellulose is the most abundant and renewable natural polymer globally and is very useful in the preparation of many materials. Cellulose consists of D-glucose monomers. These monomers are linked together by glycosidic oxygen bridges of sugar rings. Since it is a biodegradable material, there are numerous potential application zones, for example, cellulose-based materials, food bundling, biomedical, and biomaterials [6, 7]. Periodate oxidation of cellulose in cellulose repeating D-glucose units that can break the C2-C3 bond and capable of form 2,3-dialdehyde groups without significant side-reactions are highly specific reactions [8] (Fig. 1).

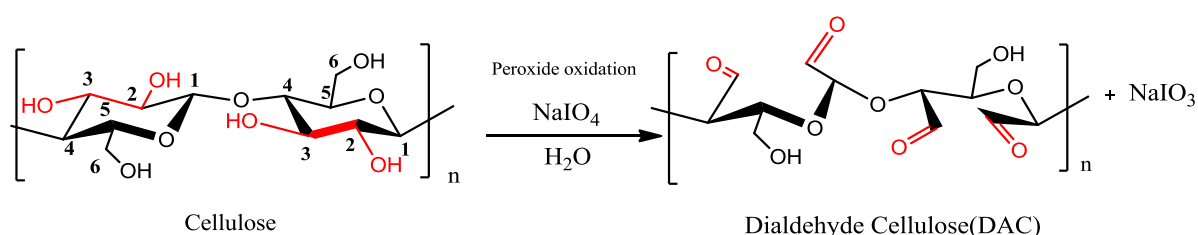


Figure 1. Reaction of cellulose with sodium periodate to produce dialdehyde cellulose (DAC).

Although hemiacetals, hemialdals and aldehyde hydrates are here, only aldehyde functions are presented to aid understanding. Since aldehyde groups are known to be easily converted to carboxylic groups, primary alcohols, and imines under certain conditions. Dialdehyde cellulose; is used to obtain cellulose-based materials for different applications such as drug carriers, heavy metal adsorbents, stabilizers for proteins, carriers for antibodies, and tissue engineering scaffolds. Cellulose aldehyde derivatives can be used as intermediates to make a variety of products such as diamines, dioximes, and Schiff bases [8-11].

Poly(lactic acid) (PLA) or polylactide is a bioplastic made from lactic acid (Fig. 2). The name "poly(lactic acid)" does not conform to the IUPAC standard designation because it is not an acid. For this reason, it can sometimes be confusing. PLA is not a polyacid (polyelectrolyte) but a polyester. PLA was discovered by Wallace Carothers, who invented nylon in the 1920s. However, the commercial use of PLA was very costly. Dr. Patrick R. Gruber and his wife Sally discovered the way to obtain PLA from corn in 1989. Thus, the production cost of PLA has decreased. In 2010, PLA was considered the second most crucial bioplastic [12-15].

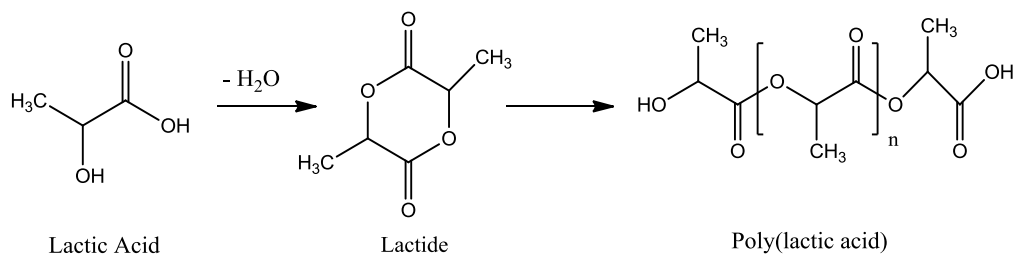
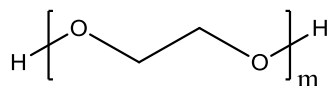


Figure 2. Reaction schemes to produce PLA

Biodegradability, upgradeable biocompatibilities, versatile physical and mechanical properties are often used in different fields, for example, PLA and copolymer, tissue layout, polymer making, soothing transport sketch, and a few clinical vitals [16]. PLA is very fragile and is not compatible with many packaging production processes. Therefore, it is necessary to strengthen it with additives. Due to their biodegradability and biocompatibility, it is FDA approved for use in medical devices [17]. Therefore, PLA finds use as a filament in 3D printing to create 3D printed parts. However, it has shortcomings such as brittleness, low impact strength, and low ability to resist thermal deformation, limiting the comprehensive application of pure PLA [18].

PLA and DAC are two promising candidates for biodegradable polymer blends [14, 15, 19]. To minimize the disadvantageous properties of PLA, it should be reinforced with natural polymer materials such as starch, cellulose, chitosan, and the like. It must be further strengthened to modify PLA materials. Blending PLA with a flexible or elastic polymer can be an alternative method to improve tensile toughness and impact toughness significantly [20].

Polyethylene glycol (PEG) (Fig. 3) is a non-circular polymer in which oxygen and ethylene units are linked sequentially. PEG is a type of polymer that is frequently preferred in medical uses. Some of the reasons for this are; non-toxic structure, hydrophilic, non-antigenicity, and immunogenicity. PEG, a biodegradable polymer, is preferred to improve mechanical properties [21, 22].



Polyethylene Glycol(PEG)

Figure 3. Chemical formula for PEG

According to Ou et al., PEG can improve the intermolecular interaction and then increase the interfacial interaction between PLA and cellulose. Because C-O-C and terminal OH can form hydrogen-bonding or dipolar interactions between PLA and the units that make up the cellulose, which can form a bridge between PLA and cellulose. Besides, they noted that adding PEG to the PLA and cellulose nanofibrils matrix showed that the tensile strength and elongation ratio increased by 56.7% and 60% compared to PLA/cellulose composites [19].

Despite the many advantages of PLA, it is still far from replacing conventional plastics due to some disadvantages. For example, since the glass transition temperature of PLA is around 60 °C, it reduces the energy requirement during processing, but it shows a weak thermal resistance. For this reason, the shape stability during processing is also poor. One way to reduce the disadvantages of PLA and improve its properties is to blend it with other biopolymers to improve its properties without losing its biodegradability. As it is known, the most abundant natural polymer found on Earth is cellulose. It has been noted that varieties such as nanocellulose, microcrystalline cellulose and cellulose fibers are used to improve the properties of PLA [23]. DAC, a modified cellulose

derivative, is a remarkable cellulose derivative with a number of interesting applications such as protein immobilization, drug delivery and its use in tissue repair or as a packaging material [24, 25].

Syama and co-workers researched the mechanical properties of composites obtained by oxidizing cellulose fibers obtained from palm fruits to dialdehyde cellulose and copolymerize them with PLA in different periods. As a result of the study, they stated that as the mixing time increased (from 1 hour to 6 hours), the tensile strength of PLA decreased and the crystallinity percentage increased. [26].

According to these studies, it is seen that the mechanical properties of PLA have been improved by using cellulose and its derivatives [23-26]. There are no studies on DAC and PLA blends prepared using the solvent technique. In the light of the previous studies, we oxidized commercially purchased microcrystalline cellulose to DAC with NaIO_4 . Then we used PEG to enable the interaction between the synthesized DAC and commercially purchased PLA [17, 27]. Finally, we characterized the obtained blends with infrared spectroscopy technique, SEM and thermal analysis techniques.

2. Experimental Methods

2.1. Materials

Microcrystalline cellulose (Alfa Aesar, degree of polymerization: 216), sodium metaperiodate (NaIO_4) Sigma-Aldrich, polyethylene glycol (Merck 6000), ethanol (Merck), ethylene glycol (Merck). The PLA used in this work was a commercial-grade type Ingeo™ Biopolymer 4043D (MFR=2,16 kg/10 min at 210 °C provided by Nature Works (Minnetonka, MN, USA).

2.2. Methods

2.2.1. Synthesis of DAC

Microcrystalline cellulose (20 g) was mixed with NaIO_4 (40 g) and 500 mL of distilled water was added (Fig. 4). After stirring the solution for 72 hours in the dark and at room temperature (25 °C), the reaction was finished by adding 16 mL of ethylene glycol. Centrifugation was carried out, and the solid was washed five times with distilled water. DAC was obtained as a white solid and dried by lyophilization.

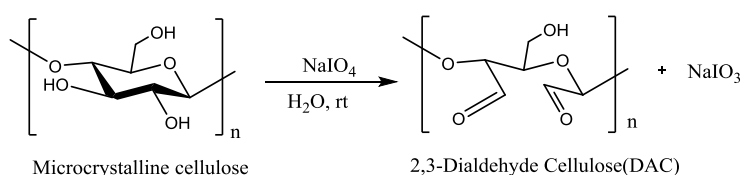


Figure 4. Experimental conditions for DAC

2.2.2. Preparation of DAC/PLA Blend

0.25 g PLA was added onto 10 mL DMF and mixed at 80 °C until the PLA dissolved. The reaction was carried out at 80 °C by adding 0.25 g of DAC to the resulting solution. The solution obtained as a result of the reaction was poured into ethanol. The liquid mixture was removed at 75° C. The obtaining blend was kept to dry under room conditions.

2.2.3. Preparation of DAC/PLA/PEG Blend

0.25 g PLA was added onto 10 mL DMF and mixed at 80 °C until the PLA dissolved. The reaction was carried out at 80 °C by adding 0.25 g of DAC and 0.025 g of PEG to the resulting solution. The resulting solution was poured into ethanol, and the liquid mixture was removed at 75 °C by taking it into the evaporator system. The resulting structure was dried under room conditions.

3. Results and Discussion

3.1. FT-IR Spectroscopic Analysis

FT-IR spectra of the samples were obtained from discs containing ~2,0 mg sample in ~20 mg potassium bromide (KBr). The measurements were carried out on a Perkin Elmer Spectrum BX FT-IR spectrophotometer at the resolution of 4 cm⁻¹ in the wavenumber region 400–4000 cm⁻¹.

3.1.1. FT-IR Spectrum of DAC and Microcrystalline Cellulose

FT-IR analysis showed that the chemical structure of DAC was significantly different from that of microcrystalline cellulose (Fig. 5). C-O-C bonds as represented by C-O vibrations at 800-1030 cm⁻¹. In the 1736 and 886 cm⁻¹ region, two specific FT-IR peaks are seen as a result of the structure of DAC. While the absorption band at 1736 cm⁻¹ belongs to the characteristic aldehyde carbonyl groups, the 882 cm⁻¹ band corresponds to the formation of hemiacetal bonds between the newly obtained aldehyde groups and their adjacent hydroxyl groups. The peak at 1632 cm⁻¹ belongs to the -OH bending of the adsorbed water because it is difficult to remove the water adsorbed in cellulose molecules completely [8, 28, 29].

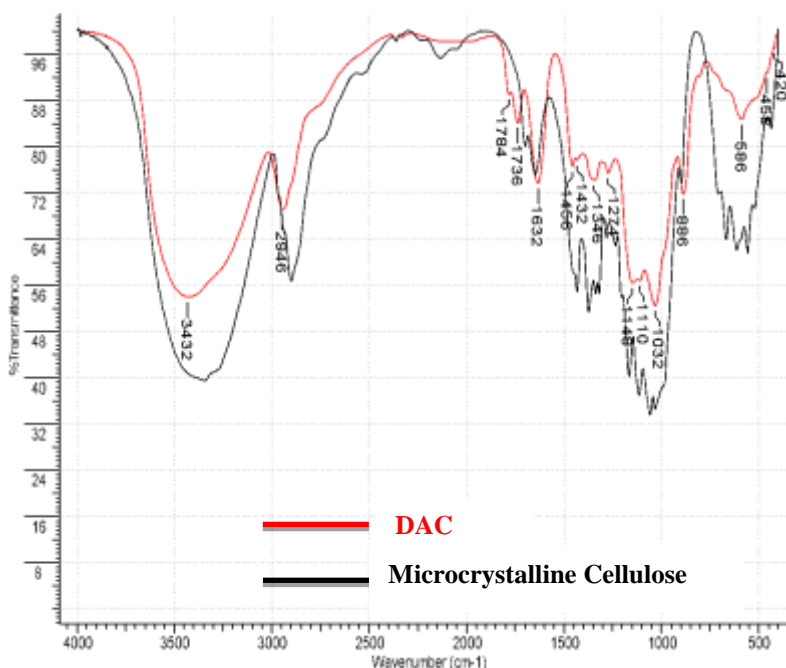


Figure 5. FT-IR spectrum of DAC and microcrystalline cellulose.

3.1.2. FT-IR Spectrum of PLA

Fig. 6 shows FT-IR spectra of PLA. PLA shows characteristic stretching frequencies for C=O, -CH₃ asymmetric, -CH₃ symmetric, and C-O, at 1756, 2946, 2996 and 1090 cm⁻¹, respectively.

Bending frequencies for $-CH_3$ asymmetric and $-CH_3$ symmetric have been identified at 1454 and 1362 cm^{-1} , respectively [30].



Figure 6. FT-IR spectrum of PLA

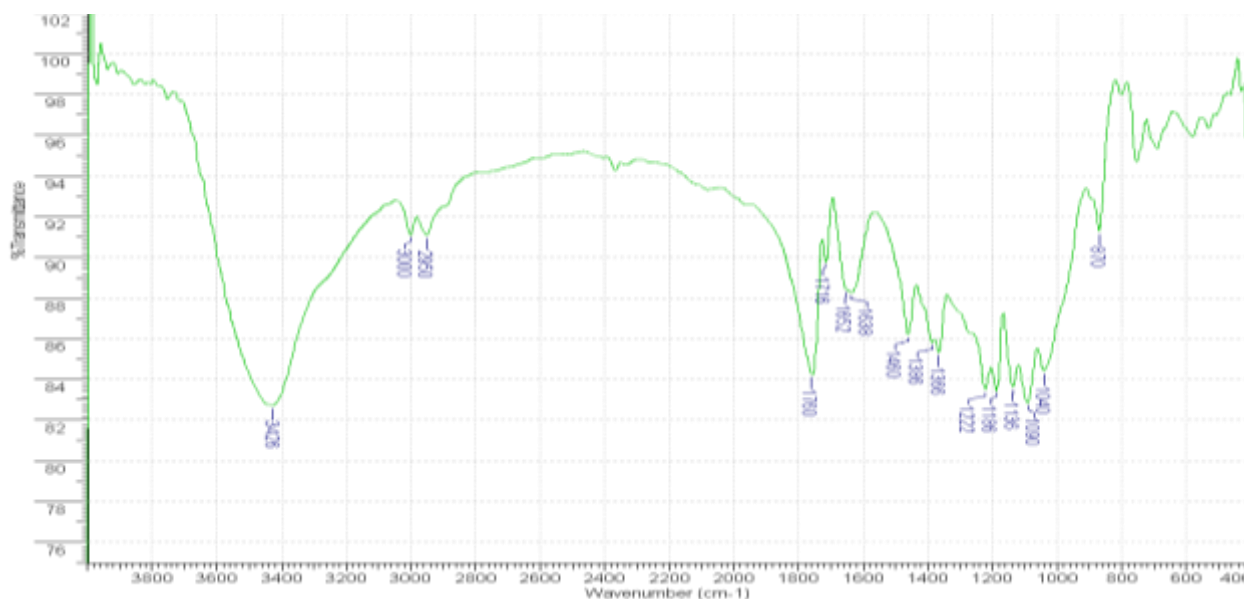


Figure 7. FT-IR spectrum of DAC/PLA blend

3.2. Thermogravimetric Analysis

The operating temperature range of the Perkin Elmer SII 7300 brand device is $25-1500\text{ }^{\circ}\text{C}$ and the heating rate varies between $0.01-100\text{ }^{\circ}\text{C}/\text{min}$. In the study, analysis was performed at a speed of 6.5

°C/min. The change of weight percent and its derivative (DTG) was recorded as a function of temperature.

3.2.1 DTG Analysis of DCA/PLA Blends

When the thermal analysis results are examined (Fig. 8a - 8b), it is seen that both polymer blends are a two-step reaction. It is observed that the blend obtained with the addition of PEG left about 12 % residue at the end of the analysis, and the deterioration in the second step started at about 285 °C.

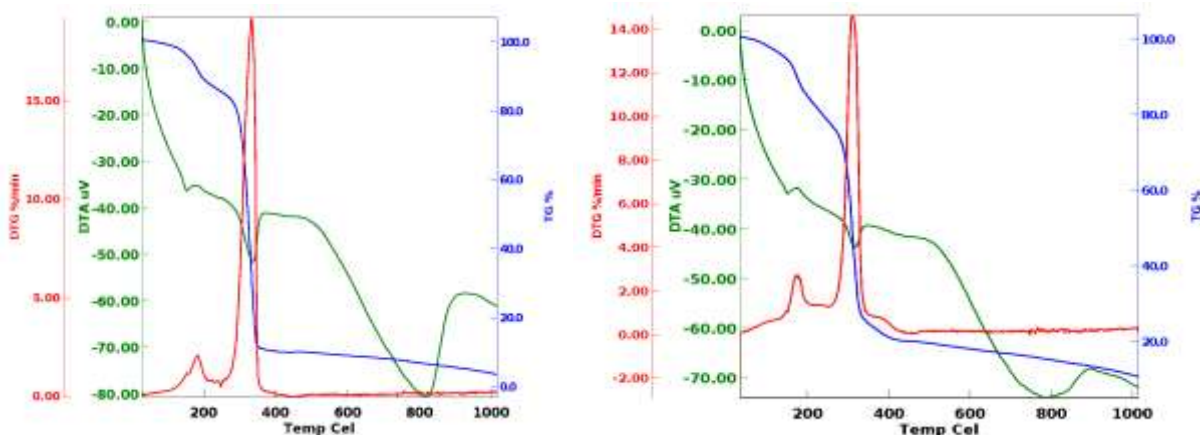


Figure 8. a-) Thermogram of DAC/PLA blend b) Thermogram of DAC/PLA/PEG blend

3.2.2 TGA Analysis of DCA/PLA Blends

The % mass-temperature change graph of the two compositions obtained in Fig. 9 is given. According to the results, it is seen that the residual rate of the DAC/PLA/PEG blend is high. Both compositions consisted of a two-step reaction. In the second step, mass change in the DAC/PLA composition graph started to happen at ~280 °C. The residue rate is seen to be around 5%. In the graph of the DAC/PLA/PEG, the 2nd step mass change started at 270 °C, and the residue ratio is observed as 12%.

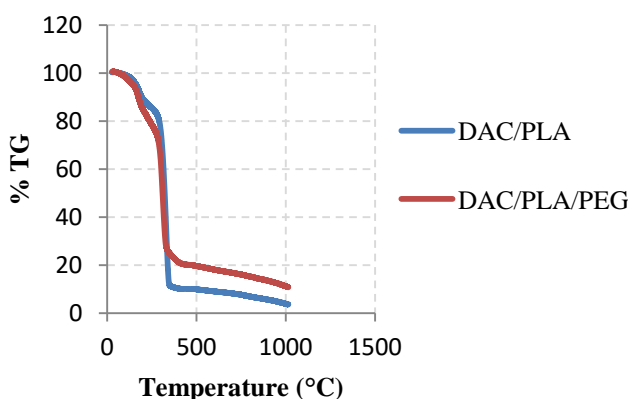


Figure 9. Comparative % mass-temperature change

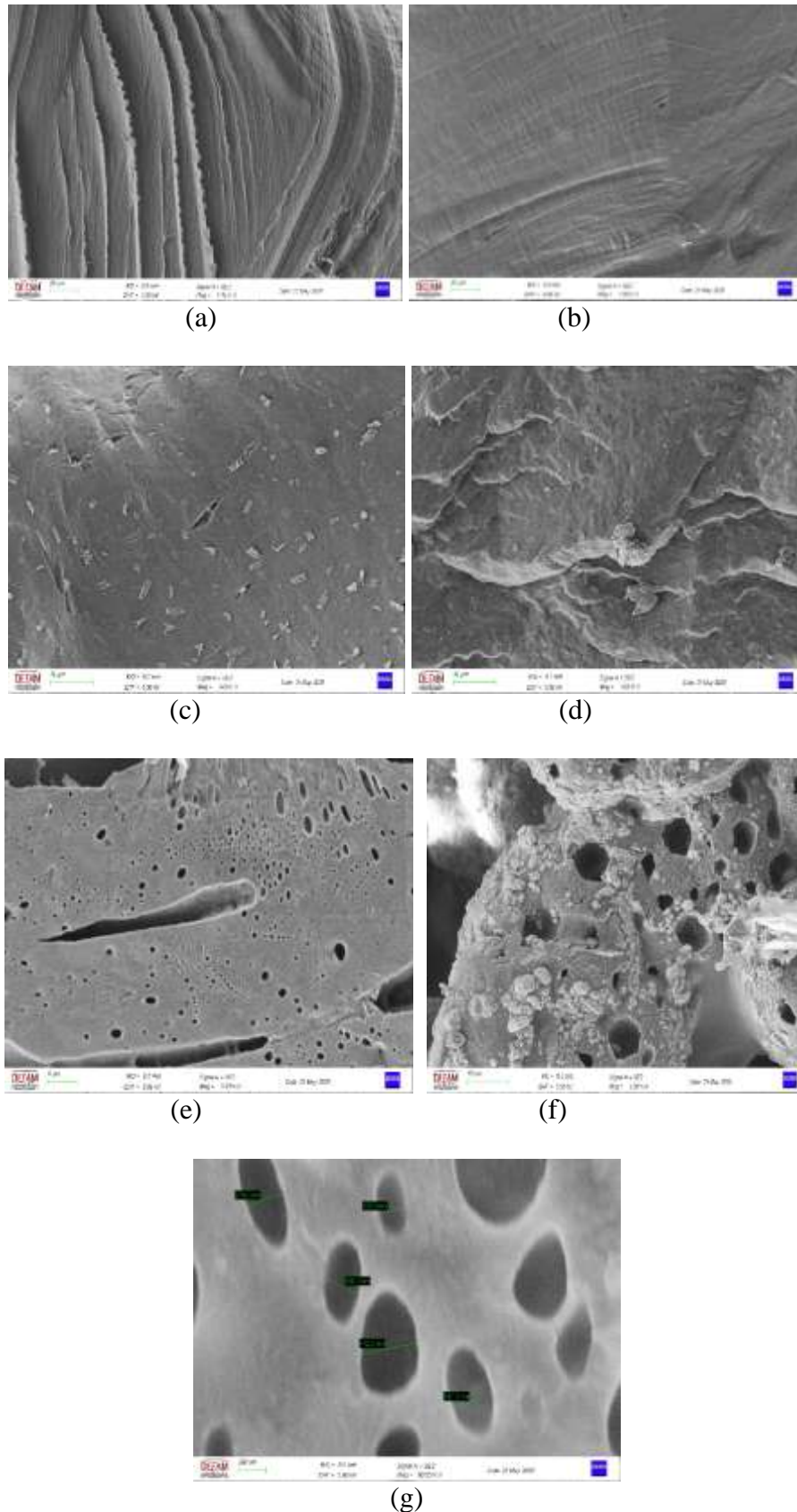


Figure 10. a-b) SEM image of DCA c-d) DAC/PLA blend e-f-g) DAC/PLA/PEG blend

It is concluded that the polymer blend degradation having DAC/PLA/PEG composition in the first step starts at a lower temperature. According to these results, it can be said that the first structure to deteriorate in the structure is PEG.

3.3. Morphological Analysis of Blends

The surface morphology of DAC/PLA and DAC/PLA/PEG blends were investigated using Zeiss Gemini 500 field emission scanning electron microscopy (SEM) at Manisa Celal Bayar University (DEFAM). The prepared samples were characterized using a field-emission scanning electron microscope operating at 10 kV under a high vacuum.

The fracture surface morphology of PLA/DAC and the DAC/PLA/PEG blends are showed in Fig. 10. It is seen that there are fibrous structures. DAC appears to have fibrous structures. Although these fibers are not seen in all regions, they are not regular (Fig. 10a-10b). It can be seen that there is no interaction between the apolar and polar groups (Fig. 10c-10d). As a result of the interaction with the addition of PEG in the blend, it is seen that there are many holes in the new blend (Fig. 10e-10f). It was determined that the diameters of these holes were between 205-412 nm (Fig. 10g). Also, it is seen that the fibrous structure changes and rounding occur with the addition of PEG. It can be said that the SEM images obtained are compatible with the papers of Qu and coworker's that about the properties of the blends prepared with PLA and PEG [31].

In this study, PEG was used to ensure compatibility between hydrophobic PLA and hydrophilic DAC. PEG is a good compatibilizer for PLA. Hydroxyl groups of PEG interact with DAC. Another reason for the PEG function is that we want to improve the mechanical properties of the blend. PEG not only improves intermolecular interaction, but it also improves interface interaction because the C-O-C and O-H bonds act as bridges between the hydrogen bond or dipolar interactions connecting PLA and DAC (Fig. 11) [19, 32-34].

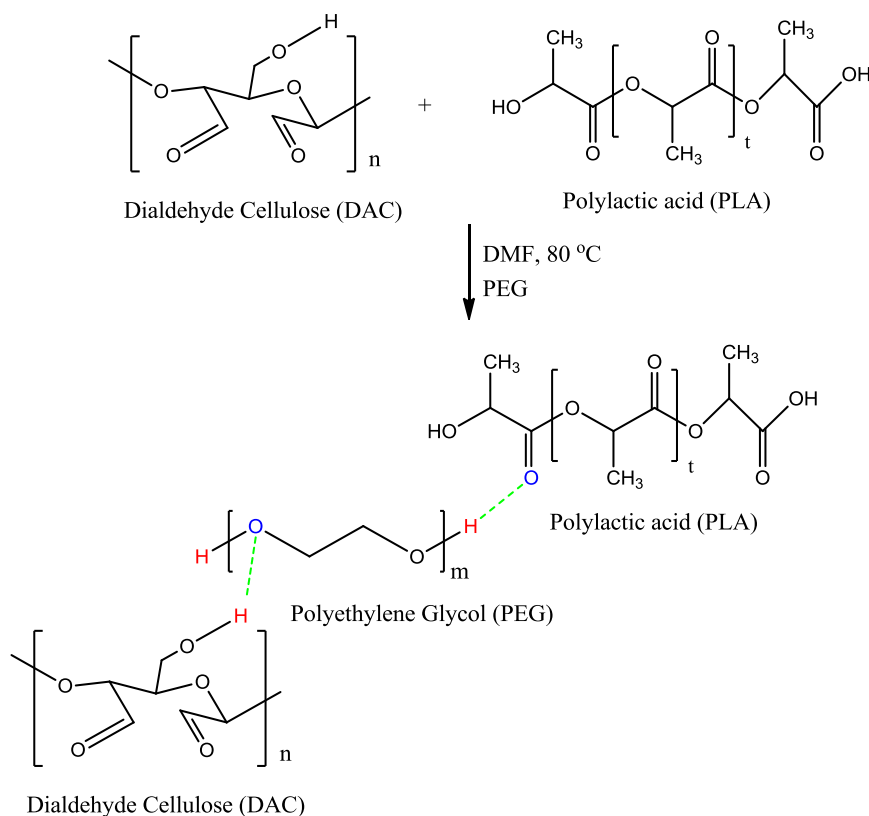


Figure 11. One of the possible interactions between DAC/PLA blend and PEG.

According to Kim et al., the mechanical properties of the immiscible polymer blends are largely dependent on the size of the dispersed areas and the interfacial adhesion between the matrix and the dispersed phase [29]. In our paper, no experimental study was conducted on the optimum amount of

PEG used as a compatibilizer and its contribution. Therefore, the mechanical properties of the blends were not investigated. However, there are a few studies on cellulose-based blends and the addition of PLA to improve thermal and mechanical properties [35-37].

3. Conclusions

In this study, it was aimed to obtain a new polymer blend based on cellulose and PLA, natural biodegradable polymers. The solvent technique was used for blending method, and PEG was preferred as a compatibilizer between DAC and PLA. Thermal analysis of the blends obtained were carried out. It was observed that the residual mass amount increased but the decomposition temperature decreased with the use of PEG as compatibilizer. Besides, possible interactions between structures were shown with SEM images. According to related literatures, it can be said that the harmony between the multiple holes DAC/PLA formed in SEM images with the addition of PEG is achieved. Functional groups were determined with FT-IR spectra of DCA, PLA and DAC/PLA blend. Finally, although we have examined properties of the DAC and PLA blend with some techniques, extensive studies are also required to determine the mechanical properties and biodegradation processes of the blends. Future studies can be carried out by determining the optimum blending ratios.

Authors' Contributions

This study includes the results of ÜGS's master thesis. KŞ designed the synthesis studies of the project. ÜGS performed the experimental procedure. EA followed the synthesis steps and interpreted the analysis results of the obtained polymers. EA and KŞ evaluated the results in cooperation and wrote the article. All authors have read and approved the final version of the manuscript.

Competing Interests

The authors declare that they have no competing interests.

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