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Immobilization of propolis extract on PET fabric for biomedical applications

Biyomedikal uygulamalar için propolis ekstresinin PET kumaş üzerine immobilizasyonu

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Immobilization of Propolis Extract on PET Fabric for Biomedical Applications

Highlights

- *Obtaining flavonoids from propolis by ethanolic extraction*
- *Modification of PET fabric with ethylenediamine*
- *A technical textile preparation by immobilization of propolis extract onto the PET fabric*
- *Chemical, physical and mechanical characterization of the prepared technical textile*
- *Antibacterial activity of the technical textile*

Graphical Abstract

Flavonoids, which are bioactive materials from propolis, were immobilized on chemically modified PET fabric by using a crosslinking agent. FT-IR spectroscopy, contact angle measurements, optical microscope images, mechanical tests, and antibacterial tests were performed for characterization of the fabric.

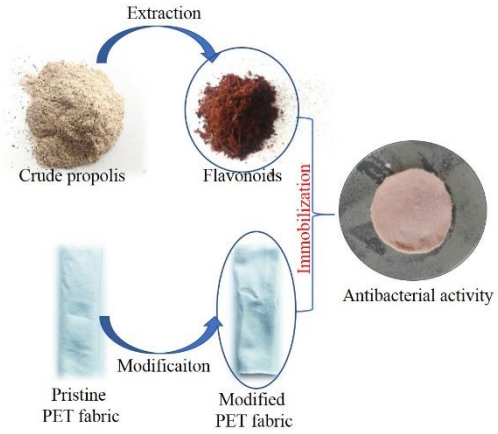

Figure. Modification of PET fabric, extraction of propolis, and the immobilization of propolis extract on PET fabric

Aim

Preparation of a technical textile for biomedical applications from PET fabric and propolis

Design & Methodology

PET fabric was chemically modified and the flavonoids from propolis were immobilized.

Originality

PET fabric was functionalized with propolis.

Findings

FT-IR spectrum and water contact angle measurements prove the modification and immobilization. An antibacterial activity of the fabric against gram negative bacteria was determined.

Conclusion

A new technical textile from natural materials for biomedical applications was developed.

Declaration of Ethical Standards

The authors of this article declare that the materials and methods used in this study do not require ethical committee permission and/or legal-special permission.

Immobilization of Propolis Extract on PET Fabric for Biomedical Applications

Araştırma Makalesi / Research Article

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ABSTRACT

Propolis has biological activity due to its wide variety of phenolic compound content. Propolis usage in many areas such as cosmetics, food, medicine, and biomedicals is becoming widespread day by day due to its antibacterial, antifungal, antiviral properties. In this study, phenolics extracted from propolis were covalently bonded and immobilized on the surface of polyethylene terephthalate (PET) fabric with the aim of developing a technical textile for biomedical applications. To do so, PET fabric was aminated, and then the phenolics were immobilized using polyethylene glycol diglycidyl ether (PEGDGE) as the crosslinking agent. Formation of amine groups in PET structure and immobilization of the phenolics were proved by ATR-FTIR spectrums. Water contact angle of PET being 121° decreased to 110° and 97° after the amination and the immobilization, respectively. Optical microscope images were taken to monitor morphological changes after the processes. DSC results revealed a new endothermic peak at around 40 °C for modified PET. Tensile tests showed that tensile strength of the fabric weakens upon modification, while a healing effect occurs during immobilization. Antibacterial tests revealed that propolis extract immobilized fabric has an antibacterial activity against gram negative (*E. coli*) bacteria.

Anahtar Kelimeler: Propolis, polyethylene terephthalate, immobilization, technical textile, antibacterial activity.

Biyomedikal Uygulamalar için Propolis Ekstresinin PET Kumaş Üzerine İmmobilizasyonu

ÖZ

Propolis, çok çeşitli fenolik bileşik içeriği nedeniyle biyolojik aktiviteye sahiptir. Propolisin antibakteriyel, antifungal, antiviral özellikleri nedeniyle kozmetik, gıda, ilaç, biyomedikal gibi birçok alanda kullanımı her geçen gün yaygınlaşmaktadır. Bu çalışmada, biyomedikal uygulamalar için teknik bir tekstil geliştirmek amacıyla propolisten ekstrakte edilen fenolikler polietilen tereftalat (PET) kumaş yüzeyine kovalent olarak bağlanmış ve immobilize edilmiştir. Bunu yapmak için, PET kumaş aminlendi ve daha sonra fenolikler, polietilen glikol diglisidil eter (PEGDGE) çapraz bağlama ajanı kullanılarak kumaş üzerin immobilize edildi. PET yapısında amin gruplarının oluşumu ve fenoliklerin immobilizasyonu ATR-FTIR spektrumları ile kanıtlanmıştır. 121° olan PET'in su temas açısı aminasyon ve immobilizasyon sonrasında sırasıyla 110° ve 97°'ye düşmüştür. İşlemlerden sonraki morfolojik değişiklikleri izlemek için optik mikroskop görüntüleri alındı. DSC sonuçları, modifiye edilmiş PET için yaklaşık 40 °C'de yeni bir endotermik pik ortaya çıkardı. Çekme testleri, kumaşın mukavemetinin modifikasyon üzerine zayıfladığını, immobilizasyon sırasında ise onarıcı bir etki meydana geldiğini göstermiştir. Antibakteriyel testler, propolis ekstresi immobilize edilmiş kumaşın gram negatif (*E. coli*) bakterilere karşı antibakteriyel aktiviteye sahip olduğunu ortaya koymuştur.

Anahtar Kelimeler: Propolis, polietilen tereftalat, immobilizasyon, teknik tekstil, antibakteriyel aktivite

1. INTRODUCTION

Propolis, often called ’bee glue’ is known as a variety of plants collected by honeybees such as poplar, palm, conifer secretes, pine, gums, leaf buds, mucilage, and resins. It is used as an antiseptic in the construction, sealing and protection of cracks in the honey bee hive against microbial infections, preventing the decomposition of intruders, and maintaining internal temperature [1]. The physical properties and chemical structure of propolis vary depending on geographical origin, the season in which it is collected, and the type of plant source [2]. Its color varies between green, brown and red [3]. It is a sticky natural substance and melts at 60 -70 °C, while some propolis melt at around 100 °C [4]. Propolis is a mixture of essential oils and decks, and it is also a natural substance containing minerals, amino acids, vitamins, and flavonoids [5]. Propolis has extensive biological activities namely anti-inflammatory, anti-oxidant [6], anti-viral [7], anti-bacterial [8], anesthetic, anti-septic [9], anti-cancer [10], anti-hepatotoxic [11], anti-fungal [12], anti-tumoral [13], anti-mutagenic [14].

The chemical component of propolis is very complex, and around 300 compounds have been determined in its composition. Its chemical properties also depend on the geographical diversity of plant resources and bee species [15]. The chemical composition of propolis, especially its phenolic profile, is affected by deciduous plant sources. In the temperate zone, propolis is identified with low content of phenolic acids and esters and high content of

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flavanones and flavones. Phenolic compounds having more complex profiles have been identified as caffeoylquinic acid derivatives, prenylated p-coumaric acids, lignans, and prenylated flavonoids [16]. These bioactive compounds have been extensively extracted in order to evaluate in biomaterials. Different solvents are used to obtain rich propolis extracts such as polyphenolic components; wetting is performed by reflux, shaking, or Soxhlet extraction [17]. The most common solvent for the preparation of propolis extracts is absolute ethanol [18].

In many applications of propolis extracts in biomaterials, generally polymer substrates are used. For instance, Wo´zniak and coworkers impregnated propolis extract into wood by using a silane agent in order to preserve creep performance of the wood in long term. They also offered to use this bio-friendly product in various biomedical applications [19]. In another study, Reyes et al. loaded red propolis ethanolic extract (RPEE) into gelatin-based films to gain antibacterial and antioxidant properties. From the biological test, they found high antioxidant activity and antibacterial property against the Gram(+) bacteria [20]. Khodabakhshi and coworkers coated polyurethane (PU) foams with propolis with the aim of using in dressing application. They reported enhanced in vitro cellular compatibility and in vivo wound healing activity with increasing coated propolis concentration [21]. Torlak and Sert coated polypropylene film with chitosan–propolis and investigated antibacterial effectiveness against foodborne pathogens. It was concluded that incorporating ethanolic extract of propolis (EEP) enhances antibacterial and antimicrobial activity at 10% EEP/chitosan content against all pathogens tested. They suggested that this film can be used in food packaging applications. [8].

In literature, there are a few studies on the integration of propolis extracts into fabrics. In one of these studies, Rogina-Car et al. prepared EEP containing Tencel® nonwoven fabrics by immersing and scattering procedures for wound care application. They concluded that, EEP treatments on nonwoven fabrics provide significant microbial barrier effect [22]. In another study, propolis extract was encapsulated and loaded into cotton fabric by Sharaf and El-Neggar with the aim of obtaining a wound dressing material [23]. Turan et al. coated cotton fabrics with propolis loaded microcapsules and demonstrated antibacterial activity against gram negative and gram positive bacteria [24]. In other studies, propolis was impregnated on cotton fabric [25-28]. From the literature review, it is seen that only cellulosic fabrics have been used related to propolis containing technical textiles. The reason could be exploitation of hydroxyl groups in the chemical structure of cellulose derivatives to provide an interaction with functional groups of propolis. Usage of synthetic fabrics containing propolis has not been reported so far.

Among other synthetic fabrics, polyethylene terephthalate (PET) fabric has been preferred in biomedical materials owing to its high mechanical properties, bio durability, and low cost [29, 30]. However, chemical inertness and hydrophobic character limit its usage in preparation of biomedical materials. Therefore, in order to incorporate with biologically active materials, polyester fabric requires chemical or physical modification. For instance, Zhang et al. etched polyester fabric with sodium hydroxide solution (NaOH) to form pits and embedded magnetic particles into them. Afterward, they coupled these particles with □-cyclodextrin for controlled release of menthol with the purpose of the treatment of cutaneous diseases [31]. In another study, Mrácek and coworkers firstly treated the polyester fabric surface with plasma, then grafted it with allylamine to obtain a wound healing biomedical material [32]. Joseph and Rajeev coated woven PET fabric with polyvinylidene fluoride and evaluated it in possible usage as sewing ring of prosthetic heart valves.

Owing to its very rich and diverse phenolic content, propolis has been used in many biomedical applications such as cell regenerative, antibacterial, antiviral, and anti-aging. However, there has been no study on the integration of propolis components into PET fabric. The aim of this study is the immobilization of phenolic components of propolis on PET fabric in order to develop a new biomaterial. This new material can be a candidate for various applications such as wound healing bandages thanks to its moisture retention properties and surgical yarn thanks to cell regeneration feature. Hence, a new technical textile has been introduced and a high value-added product has been developed.

2. MATERIAL and METHOD

2.1. Materials

Pure propolis powder, which is harvested from central Anatolia, was purchased from Zencefil Organik®, Turkiye. Knitted 100% PET fabric was kindly supplied by Yesim Tekstil, Turkiye. Ethanol absolute (≥ 99.9) was supplied from ISOLAB chemicals, poly(ethylene glycol) diglycidyl ether (average Mn 500) was supplied from Sigma-Aldrich, and ethylenediamine (for synthesis) was supplied from Merck.

2.2. Extraction of Propolis

Phenolic components in propolis composition were extracted using ethanol. To do so, 10 grams of raw propolis were added into 100 mL of 30% ethanol solution in a tightly closed glass bottle. Then the mixture was ultrasonicated for 5 min in an ultrasonic bath (HY Teknoloji Hy-6 D, Turkiye). Afterward, it was stirred at 1000 rpm for 1 hour at 25 °C on a magnetic stirrer (Heidolph, Hei-Tec, Germany). The mixture was filtered, and the filtrate was collected. 3.96 grams of the ethanol extract of propolis (EEP) was obtained by vaporization of the solvent and dried under vacuum (Jeiotechi, Korea) at 60 °C for 24 hours.

2.3. Modification of PET Fabric

PET fabric was modified by amination. Typically, a piece of PET fabric in dimensions of 5 cm x 15 cm was

taken to a mixture of 25 mL ethylenediamine (EDA) and 75 mL distilled water in a tightly closed glass bottle containing a magnetic bar. Then the bottle was placed in a water bath and stirred at 100 °C for 30 minutes on the hot plate. Then, the heater was turned off, the temperature was allowed to decrease to 50 °C and the modified PET (MPET) was taken out (Figure 1). MPET thoroughly washed with distilled water and dried at 75°C in an oven (MMM group Ecocell, Germany).

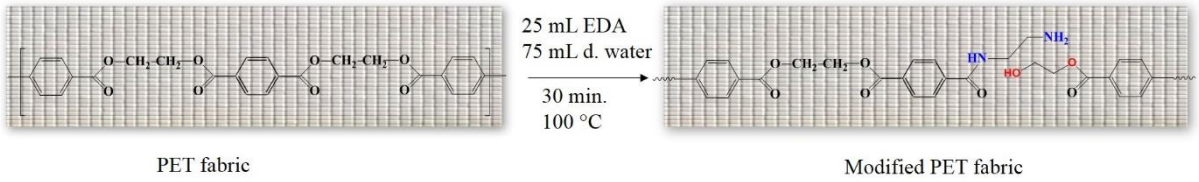

Figure 1. Modification of PET fabric.

2.4. Immobilization of EPP on MPET Fabric

Immobilization of EEP was carried out by utilizing amine groups of MPET. Poly(ethylene glycol) diglycidyl ether (PEGDGE) was used as crosslinking agent between MPET and EEP. Firstly, 1 gram of EEP was added to 75 ml tetrahydrofuran (THF), which is inert against the reactants, and stirred for 5 min. Then MPET with 5 cm x15 cm dimensions was taken into the EEP solution. Subsequently, 4 ml of PEGDGE was added dropwise. The reaction mixture was stirred for 1 hour at 25 °C. The EEP immobilized PET fabric (EEP-PET) was washed with excess amount of distilled water and dried at 75°C after for 24 hours. The procedure was depicted in Figure 2.

2.5. Characterizations and Tests

Chemical structures of all the samples were investigated by attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FTIR) using Thermo Scientific Nicolet iS50 FTIR Spectrometer (USA). The results were recorded a mean of 16 repeating scans in the wavelength range of 4000-400 cm- 1 with a resolution of 1 cm-1.

Differential scanning calorimetry (DSC) was used to determine thermal transitions of the samples using TA Instruments DSC25 (USA) calorimeter with a 10 °C/min heating speed between -70 – 300 °C under flux of nitrogen gas (50mL/min).

Contact angle (CA) measurements were carried out at room temperature using Biolin Scientific/Thetaflex optical tensiometer (Sweden).

In order to investigate any changes on the fabric surfaces, optical microscope (OM) images were taken using a stereo (up-right) microscope (Leica-M125, Germany).

Mechanical properties of the samples were determined by tensile tests using SHIMADZU-AGS-X (Japan) universal test instrument equipped with 1 kN loadcell. Tests were conducted with 5 cm x 15 cm fabrics in warp

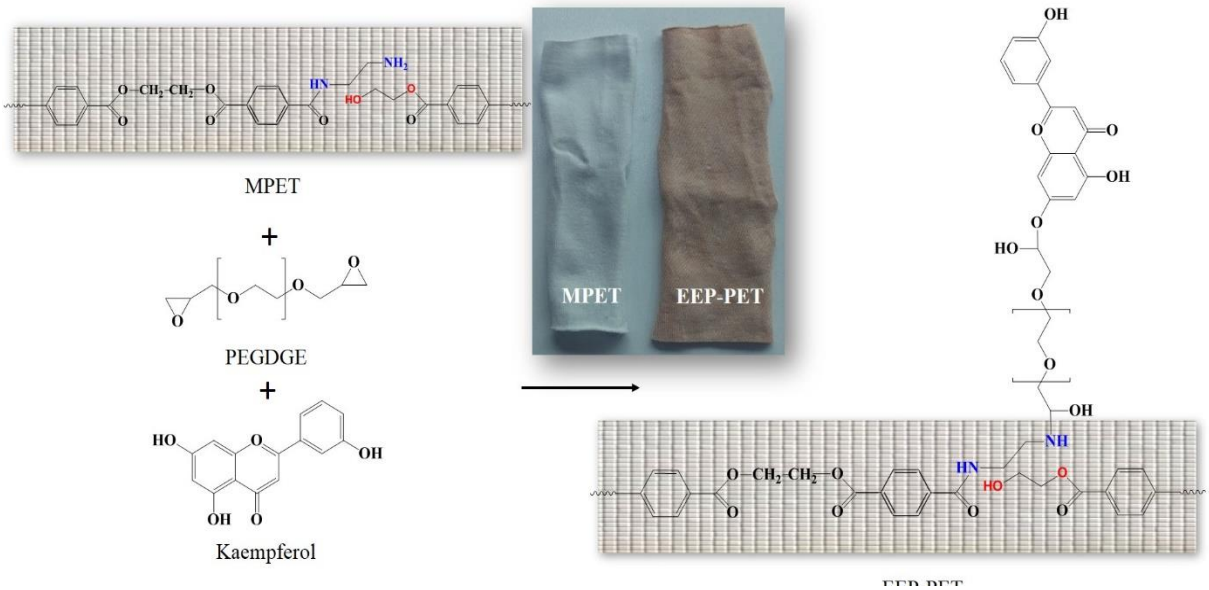

Figure 2. Immobilization of EEP on MPET (Kaempferol which is one of the EEP content was used as a representative). Inset is pictures of MPET and EEP-PET.

direction and the crosshead speed was set to 5 mm/sec. Mean value of three repetitive tests was given as the results.

Antibacterial studies were performed by agar diffusion plate test. The antibacterial activities were investigated against *Staphylococcus aureus* (*S. aureus*, ATCC 6538, G+ve) and *Escherichia coli* (*E. coli*, ATCC 10536, G−ve). *S. aureus* and *E. coli* were activated and spread on the agar. The fabrics in circular shapes were placed on the bacteria covered agar surface. The plates were incubated at 37 °C for 24 h and antibacterial activity was evaluated by observing inhibition zone formations around the fabrics.

3. RESULTS AND DISCUSSION

3.1. FT-IR Analysis

In order to observe the changes in the chemical structure of PET fabric during the modification with EDA, the FTIR spectrums were recorded (Figure 3). In the spectrum of PET, peaks at 2858 and 2956 were ascribed to aliphatic -CH_2 groups, strong peaks at 1713 cm^{-1} and 1244 cm^{-1} were attributed to the carbonyl group of ester unit. The benzene ring vibration peak appeared at 872 cm^{-1} [33]. In the spectrum of EDA, the peaks at 3363 cm^{-1} and 3287 cm^{-1} were attributed to primary amine (-NH_2) in the presence and absence of H-bonding, respectively. The peaks at 2918 cm^{-1} and 2848 cm^{-1} belong to aliphatic -CH_2 groups. The peak at 1596 cm^{-1} was ascribed to bending vibration of -NH_2, whereas the strong peak at 811 cm^{-1} was attributed to bending vibration of N-H plane [34]. The characteristic peaks of both PET and EDA were observed in the MPET spectrum. Amine peaks appeared at around 3296 cm^{-1} while carbonyl peak arising from PET was observed at 1711 cm^{-1}. The two peaks at 1647 cm^{-1} and 1549 cm^{-1} were assigned bending vibrations of secondary and primary amines, respectively. These results clearly show that the amination of PET was successfully achieved.

FT-IR spectra of MPET, EEP, and EEP-PET were comparatively given in Figure 4. EEP contains flavonoid derivatives up its weight ratio of 20%. Among the others, pinocembrin, pinobenchin, galangin, and chrysin are the most abundant derivatives. They consist of aromatic rings bearing hydroxyl and carbonyl groups. Their structures have been identified and reported in the literature [16]. In EEP spectrum, a broad peak at around 3280 cm^{-1} arises from -OH groups in the flavonoid structures. The peaks belonging to aromatic -CH groups which are expected to appear just above 3000 cm^{-1} are thought being overlapped with the broad -OH peak. The peaks at 2936 cm^{-1} and 3971 cm^{-1} are attributed to the stretching of aliphatic -CH groups in some flavonoid derivatives such as tectochrysin and pinostrobin [16]. The band at 1738 cm^{-1} is assigned to carbonyl groups from flavonoid derivatives. The peak belonging to stretching vibration of aromatic C=C appeared at 1637 cm^{-1}. The peaks at around 1367 cm^{-1} were assigned to the bending vibration of -CH_2 group [35]. A very distinctive peak at around 1010 cm^{-1} is assigned to stretching of C-O-C group. Similar results for flavonoid derivatives have been reported in the literature [36, 37].

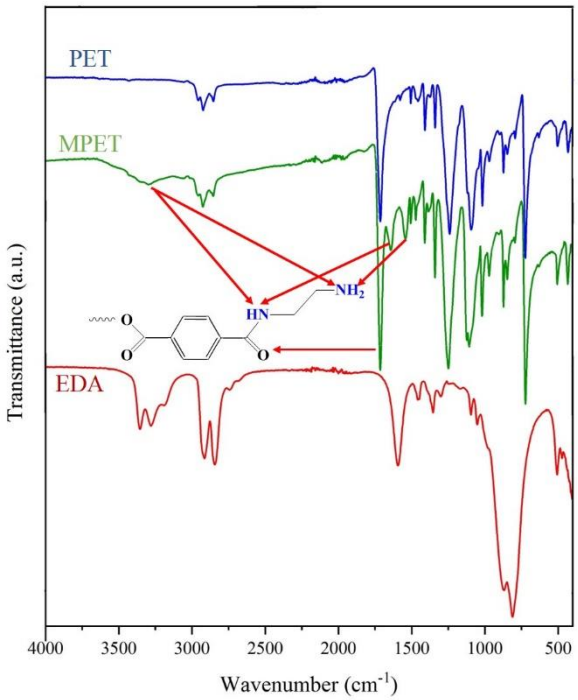

Figure 3. FTIR spectra of PET, MPET, and EDA

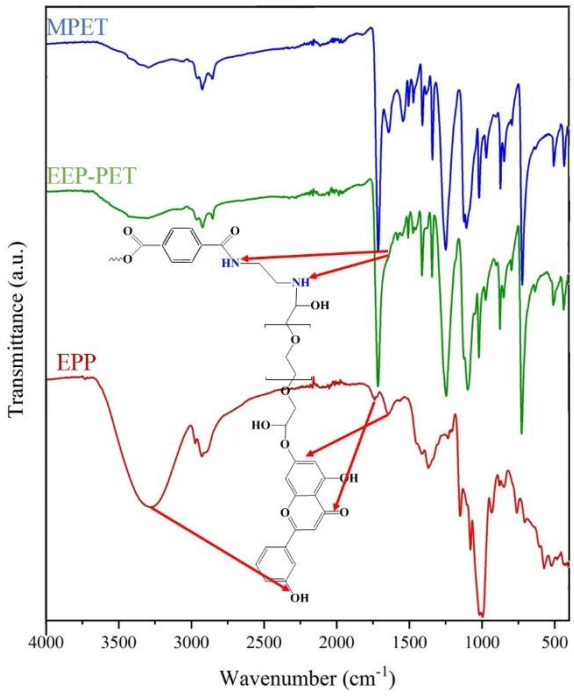

Figure 4. FT-IR spectra of MPET, EEP-PET, and EEP

In the spectrum of EEP-PET, it is clearly seen that the peak of primary amine at 1549 cm^{-1} was disappeared.

This result implies that the -NH_2 group took place in a reaction with the epoxy ring of PEGDGE. The broad shoulder at around 1638 cm^{-1} can be attributed to two different secondary amines in EEP-PET structure which can be seen in Figure 2. It is thought that the carbonyl peaks from flavonoids are overlapped with that of PET. Furthermore, when it is compared with MPET, the broadening of the peak at around 3300 cm^{-1} may be arising from -OH groups of flavonoids. These results suggest that the immobilization of EEP on PET was successfully carried out as aimed.

3.2. Contact Angle Measurements

CA measurements were conducted to observe changes in surface hydrophobicity of PET fabric upon the modification and the immobilization. As it is seen in Figure 5, raw PET fabric has a CA value of about 122° indicating a hydrophobic property. This result is in agreement with the literature. For raw PET, a CA value of 121° was reported by Liu et. al. [38]. The CA value of MPET was measured as about 110°. This decrement stems from amine groups that are formed by modification of PET with EDA. A CA value of 97° was determined for EEP-PET. This further decrement in CA is attributed to the presence of hydroxyl groups coming from the flavonoids on the fabric surface. These results demonstrate a successful amination of PET fabric and immobilization of EEP on the fabric.

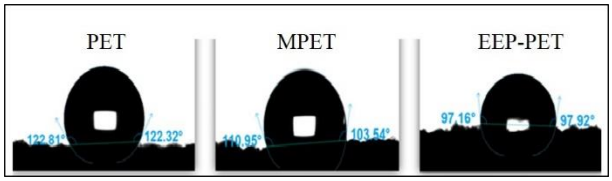

Figure 5. Water CA images of the samples

3.3. DSC Analyses

The glass transition temperatures (T_g) of polymeric materials are related to chain mobility. Chain mobility of a polymer is affected by various factors such as chemical nature, side groups, branching, and chain length. It is well known that T_g is inversely proportional to chain length and this relationship has been modeled by Flory-Fox, Beevers-White, and DiBenedetto equations. Moreover, this relation has been explained as more free volume is provided by chain ends than segments [39]. In order to observe changes in thermal transitions of PET after the modification and the immobilization, DSC curves of the samples were recorded and are depicted in Figure 6. T_g of PET was detected as 112 °C. It is well known from the literature that PET has a T_g value in the range of 75-85 °C. On the other hand, Rodrigues et al. stated that rising T_g values may be seen with increasing crystallinity as a result of stronger chain interactions [40]. Moreover, they reported varying T_g values from 105.9 °C to 171.8 °C for PET fibers having different crystallinities between 35% and 38.9%. In another study carried out by Mendes and Pereira, T_g value of 78 °C was reported for PET having 20% crystallinity [41]. By taking the melting enthalpy (DH_m) of 100% crystalline PET as 136 J/g [41], the crystallinities (X_c) of PET, MPET, and EEP-PET were calculated as 37.9, 48.7, and 45.9%, respectively. In Figure 6, similar trend with the crystallinities for T_g values of the samples were observed. These results agree with the literature mentioned above. From these findings, the high T_g values of our samples may be explained with high crystallinities. Furthermore, a new endothermic transition for MPET and EEP-PET at around 40 °C is observed. It is believed that these peaks belong to shorter chains that are formed as a result of the chain scission by EDA upon modification as depicted in Figure 1. All the samples exhibit melting at around 250 °C which is in accordance with literature [42]. The melting enthalpies were calculated as 51.5, 66.2, and 62.4 J/g for PET, MPET, and EEP-PET, respectively. The rising in the enthalpy indicates the increased amount of crystalline phase in the polymer. The crystallinity may have increased after the chemical modification as it was heated up to around T_g and then gradually cooled during this process.

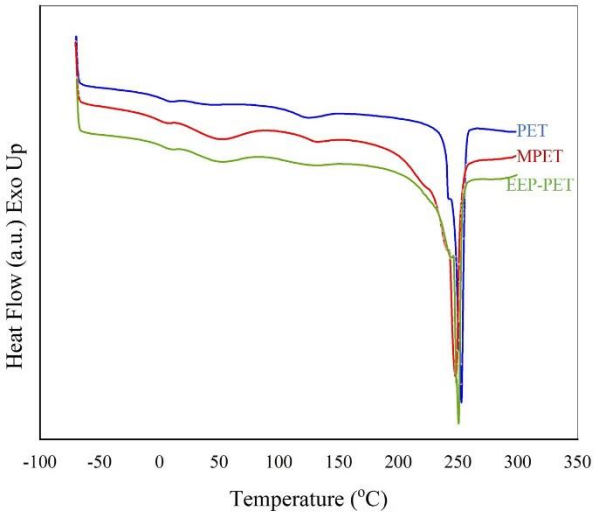

Figure 6. DSC curves of the samples

3.4. Optical Microscope (OM)OM images of the samples were recorded in order to observe any morphological changes after the amination and the immobilization processes (Figure 7). From the image of PET, a flat surface with well-ordered fibers is seen. In the images of MPET and EEP-PET, a fluffy surface with fringed and messy fibers is observed. These changes in the surface morphology of the fabric may be resulted from the chain scissions of the polymer during chemical modification which have also been revealed and discussed in DSC results.

3.5. Mechanical Test

Tensile tests have been conducted to investigate the effect of the modification and the immobilization on the mechanical properties of the fabric and the results were given in Table 1. The tensile strength of the raw fabric was determined as 13.9 MPa. Upon modification, the tensile strength dramatically dropped to 2.3 MPa which

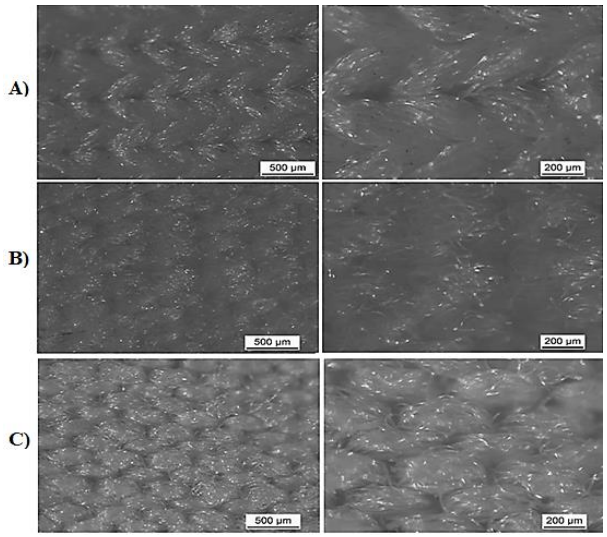

Figure 7. The optical microscope images of a) PET, b) MPET, and c) EEP-PET

is attributed to smaller chain lengths resulted from the cleavage by EDA as illustrated in Figure 1. After the immobilization, slightly increased stress for EEP-PET was measured as 3.6 MPa. This enhancement in the tensile strength suggests a healing effect that is provided most likely by PEGDGE. The healing mechanism may be explained as follows: Some of the PEGDGE molecules may link again the cleaved polymer chains that are formed during chemical modification. The strain values at break were determined as 264.3, 201.8, and 303% for PET, MPET, and EEP-PET, respectively. The lowering strain after the modification may be caused by the cleaved polymer chains, while the high strain of EEP-PET may be attributed to the introduction of PEG unit in the polymer structure as stated before.

From the stress-strain curves (Figure 8), two linear elastic regions for all samples were observed. It is thought that, the knitting texture of the fabric is responsible for the first linear region having a low slope. The slope in this region, which corresponds to elastic modulus, was determined as 0.24, 0.28, and 0.30 MPa for PET, MPET, and EEP-PET, respectively. Moreover, the strain value in this region reaches 95% for PET whereas 75% and 50% strains were observed for MPET and EEP-PET, respectively. The loss of elasticity was attributed to difficulty in sliding the fibers over each other because of the defects in the knitting texture upon the modification and the immobilization as showed with OM images. As the fabrics are stretched in the first elastic region, polymer fibers are straightened. The second region corresponds to a thermoplastic characteristic of the straight polymer fiber. Similar results were reported by Deng et al. for woven fabrics [43]. When the slopes are compared in the second viscoelastic region, a similar trend with stress values is observed. The drop in elastic modulus of MPET is due to the chain cleavages during the modification, whereas a slight increment in that of EEP-PET is again attributed to the healing effect of the PEG units. Consequently, although mechanical properties of EEP-PET are worse than pristine PET, we think that they are still enough for biomedical applications such as wound dressing.

3.6. Antibacterial Tests

Antibacterial activity of EEP-PET was investigated against *S. aureus* and *E. coli* by agar diffusion plate test. The pristine fabric as a control group was not tested since it could not be placed properly due to highly twisting behavior. It is well known already from the literature that pristine PET fabric does not have antibacterial activity against *E. coli* and *S. aureus* [44]. In the images of the

Table 1. Mechanical test results of the samples

Sample	Strength (MPa)	Elongation (%)	Elastic Modulus I (MPa)	Elastic Modulus II (MPa)
PET	13.9 ± 1.1	264.3 ± 12.5	0.25 ± 0.05	15.3 ± 1.5
MPET	2.3 ± 0.7	201.8 ± 4.5	0.28 ± 0.04	1.6 ± 0.2
EEP-PET	3.6 ± 0.5	303.1 ± 9.9	0.30 ± 0.04	3.2 ± 0.5

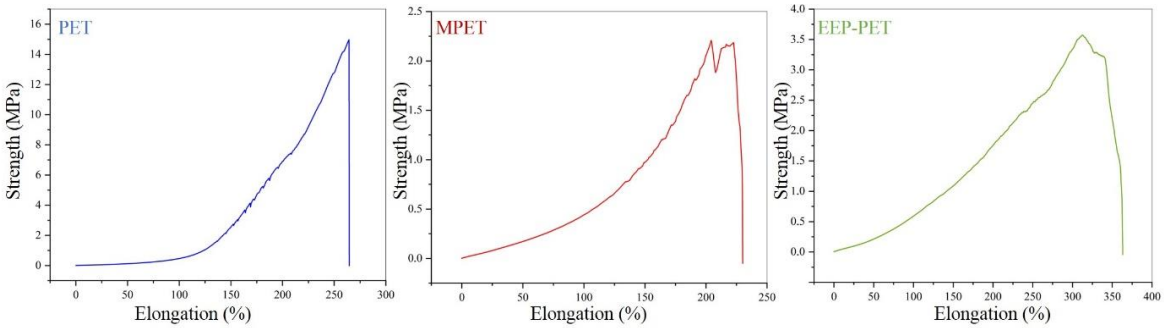

Figure 8. Tensile curves of the samples

test (Figure 9) any formation of inhibition zone was observed. However, it is obvious that *E. coli* growth did not occur on the fabric surface, whereas *S. aureus* covers the surface. This result clearly shows that the EEP-PET has an antibacterial activity against *E. Coli*. It is stated many times in the literature that, antibacterial activity of propolis against gram positive and gram negative bacteria varies depending on some factors such as the origin country of the propolis and extraction method [16]. Sharaf et al. investigated antibacterial performance of propolis induced cotton textiles. They reported larger inhibition zones with increased propolis content of cotton fabrics [26]. They explained the formation of the inhibition zone by migration and diffusion of the antibacterial agent from the fabric towards to agar media. From that point, the non-formation of inhibition zones in this study for EEP-PET is reasonable since EEP is immobilized on the fabric by a crosslinking agent which prevents the migration of the molecules of EEP.

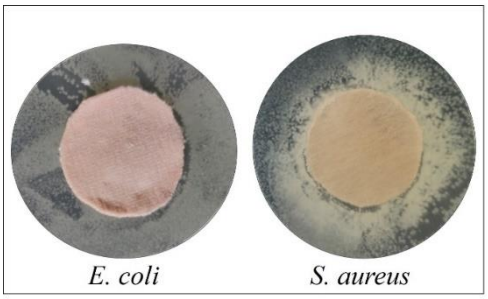

Figure 9. Antibacterial test results of EEP-PET

4. CONCLUSION

PET fabric was chemically modified in order to introduce functional groups for immobilization. Flavonoids extracted from propolis were immobilized on the MPET using PEGDGE as crosslinking agent. Both the modification and the immobilization were proved by FT-IR spectroscopy. From the CA measurements, increasing hydrophilicities were determined after the modification and the immobilization. OM images revealed the modification and the immobilization caused some defects in the fabric texture. Moreover, a new endothermic peak at around 40 °C appeared and was attributed to the shorter polymer chains resulted from the modification. Furthermore, it was determined that modification weakened the mechanical properties of the fabric. However, a healing effect after the immobilization was observed. It is thought that the mechanical properties of EEP-PET are adequate for biomedical applications. It was concluded that the EEP-PET has an antibacterial activity against gram negative (*E. coli*) bacteria. Hence, a new technical textile was developed using a natural source which has potential applications as a biomedical material.

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DECLARATION OF ETHICAL STANDARDS

The authors of this article declare that the materials and methods used in this study do not require ethical committee permission and/or legal-special permission.

AUTHORS' CONTRIBUTIONS

Omer Yunus GUMUS: Sample preparations, characterizations, testing, Writing - review & editing.

Ismahane YSSAAD: Sample preparations, characterizations, testing.

CONFLICT OF INTEREST

There is no conflict of interest in this study.

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