



Multifunctional polymer nanocomposite coated antibacterial filters for air-purification

Hava temizleme için çok fonksiyonlu polimer nanokompozit kaplamalı antibakteriyel filtreler

Arife Kübra Yontar^{1,*} , Onur Yontar² , Sinem Çevik³ 

¹ Ondokuz Mayıs University, Samsun Vocational School, Department of Mechanical and Metal Technology, 55100, Samsun, Türkiye

² Ondokuz Mayıs University, Department of Mechanical Engineering, 55139, Samsun, Türkiye

³ Ondokuz Mayıs University, Department of Metallurgical and Materials Engineering, 55139, Samsun, Türkiye

³ Ondokuz Mayıs University, Department of Nanoscience and Nanotechnology, 55139, Samsun, Türkiye

Abstract

Products used and produced in industrial locations must have natural ingredients and minimal energy usage due to air and environmental pollution. It is impossible for standard filters to clean bacteria in homes, workplaces and areas working with biological creatures. Therefore, the production of multifunctional nanocomposite and hybrid filter papers is necessary. In the study, filter papers coated with nanocomposite solutions prepared from green synthesized nanosilver (AgNPs) with plant extracts and Polyvinyl Alcohol (PVA). Scanning Electron Microscope (SEM) analyses showed that silver nanoparticles and PVA homogeneously coated the filter paper fibers, thereby reducing the average pore diameters by only 3% compared to commercial standards of filter paper. A filter with 36% higher burst pressure resistance was produced by nanosilver, plant extracts and PVA modification. The antibacterial effect is provided to the filters with a high effect of up to 10000 times, by coating them with nanosilver and plant extracts. This study made it possible to produce filter paper with natural ingredients and at low cost, reducing energy and raw material consumption, with high antibacterial effect and mechanical strength. In this way, bacteria and pollutants in the air will be cleaned without harming the environment and human health.

Keywords: Antioxidants, Air filters, Plant extracts, Green synthesis, Air purification, Nanosilver

1 Introduction

As stated by the World Health Organization (WHO), almost 90% of the global population lives where air pollution levels exceed the guidelines for air quality. Air pollution and particulate matter (PM) have become one of the most critical problems of our time. Environmental pollution, various diseases and particulates and airborne microorganisms that cause climate crisis occur with industrial applications and automobile emissions [1, 2]. The main constituents of PM include chemical impurities such as elemental carbon, sulfate, organic carbon, chloride, nitrate, iron, and calcium,

Öz

Hava ve çevre kirliliği, endüstriyel alanlarda kullanılan ve üretilen ürünlerin doğal içerikli ve düşük enerji tüketimine sahip olmasını gerektirmektedir. Evlerde, işyerlerinde ve biyolojik canlılarla çalışılan alanlarda standart olarak kullanılan filtrelerin bakterileri temizlemesi mümkün değildir. Bu nedenle çok fonksiyonlu kompozit ve hibrit filtre kağıtlarının üretimi gerekmektedir. Çalışmada yeşil sentezlenmiş nanogümüş (AgNPs), bitki ekstraktları ve Polivinil Alkol (PVA) ile hazırlanan nanokompozit solüsyonlarla kaplanmış filtre kağıtları kullanıldı. Taramalı Elektron Mikroskobu (SEM) analizleri, gümüş nanopartiküllerin ve PVA'nın filtre kağıdı elyaflarını homojen bir şekilde kapladığını, böylece ortalama gözenek çaplarını, filtre kağıdının ticari standartlarına kıyasla yalnızca %3 oranında azalttığını gösterdi. Nanogümüş, bitki özleri ve PVA kullanılarak %36 daha yüksek patlama basıncı direncine sahip bir filtre üretildi. Filtreler nanogümüş ve bitki özleri ile kaplanarak 100.000 kata kadar yüksek etki ile antibakteriyel etki sağlanmaktadır. Bu çalışma, doğal içerikli, düşük enerji ve hammadde tüketimine sahip, düşük maliyetli, antibakteriyel ve mekanik mukavemeti yüksek filtre kağıdı üretimine olanak sağladı. Bu sayede hava ortamındaki bakteri ve kirleticiler çevreye ve aynı zamanda insan sağlığına zarar vermeden temizlenmiş olacaktır.

Anahtar kelimeler: Antioksidanlar, Hava filtreleri, Bitki özleri, Yeşil sentez, Hava temizleme, Nanogümüş

as well as smoke from automobile exhaust pipes, burning coal, burning farming, and emissions from industries [3]. In addition, complex and rich pollutants in the air can come from natural sources (aerosolized soil, forest fires, pollen, dust storms) or anthropogenic (industry, transportation, construction, smoking, home heating, etc.) sources. When these pollutants are inhaled, they enter the bronchi and obstruct the airway passages and cause serious health problems including illnesses related to respiration such as severe asthma, cardiovascular morbidity, mortality, bronchitis, etc [4]. Technically, if PM is 2.5 µm or less in diameter they are clearly hazardous and, with prolonged

* Sorumlu yazar / Corresponding author, e-posta / e-mail: kubra.demirbas@omu.edu.tr (A. K. Yontar)
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exposure, can travel to the human lungs or, in certain situations, other organs in the body, this leads to increasing cardiac morbidity and respiratory. Aerosols in the air, such as volatile organic compounds (VOCs), solid particles, and bioaerosols (instance viruses, bacteria, fungus, and mold), can also cause respiratory and cardiovascular illness, as well as allergies [5, 6]. Due to these pollutants, improving air quality and providing filtration is important to protect the indoor and outdoor environment, air cleanliness and human health. The individuals spend 87% of their daily hours in their homes and 6% in cars, as indicated by the National Human Activity Pattern Survey (NHAPS). As a consequence, it is one of the most effective techniques to safeguard human health from the harmful effects of air pollution is to high-performance air purification filters are used to catch PM particles in building air conditioning systems and ventilation, as well as the use of personal face masks during outdoor activities. Air filtration technology (such as stretched electrostatic capture filtration, fiber and membrane filtration) is critical to enhancing IAQ and safeguarding human health [7]. Air Conditioning (HVAC), ventilation and heating systems in use today and all the various industrial air filters can reduce all particulate, airborne disease transmission. Because the HVAC system captures and traps both particles and microorganisms in the air filters, they can minimize microorganism rotation in the air. Bioaerosol particles grow and survive on the filter media due to dirt buildup in HVAC systems, inadequate particulate filtration levels, poor filter maintenance, or problems with moisture sources. In the long run, people indoors breathe microbial pollutants that multiply and are then released into the air, flowing through the HVAC system. As a consequence of this, major air quality issues cause health issues. Furthermore, germs are known to induce corrosion in the metallic and polymeric components of the filtering system [8]. Air filters are classified into two types: porous filters and fibrous membranes. Fiber filters have the benefit of being remarkably simple to produce on a big scale while also being energy and cost-efficient.

Traditional fiber-structured cellulose filters are currently frequently utilized in commercial air filtration applications. Other types of material used as a modification in the production of paper air filters are polyacrylonitrile (PAN), polysulfone amide (PSA), polyurethane, polyimide, polyamide 66, polylactide, polytetrafluoroethylene (PTFE), polypropylene (PP), polystyrene (PS), polyvinyl pyrrolidone (PVP), nylon-6 and biopolymer polyvinyl alcohol (PVA) polymers. PVA is a kind of synthetic polymer that is polar and biodegradable, water-soluble, inexpensive, non-toxic, has good thermal, mechanical and chemical stability, and has better environmental stability. PVA is widely used in many different kinds of applications, including artificial biomedical devices, membrane applications, electrochromic devices, paper coating, drug delivery systems, electronic devices, packaging and textile applications. In addition, PVA includes a carbon chain structure with hydroxylic groups, which can be a source for hydrogen bond interaction between nanofillers, facilitating the production of PVA-based nanocomposites [9–11]. Because of these numerous

advantages, PVA was chosen as the coating matrix material in this study.

Various yet ineffective cleaning and disinfection technologies, such as UV germicidal irradiation, air ozonolysis, and photocatalytic oxidation, have been developed to improve air quality and destroy bioaerosol particles. In recent years, various novel methods and techniques for avoiding and repelling microbial proliferation in air filters have been developed, including the use of silver nanoparticles (AgNPs), carbon nanotubes (CNT), titanium-based nanoparticles, iodine powders, and copper nanoparticles [4]. In general, because of its broad spectrum anti-pathogen effect, silver is the most extensively utilized antibacterial metal. AgNPs can kill more than 650 types of pathogens such as viruses, fungi and bacteria. Silver nanoparticles perform by several mechanisms that include metal ion release, non-oxidative and oxidative stress induction reactions are examples of such processes. The progressive transmission of silver ions released from the AgNP solution prevents bacterial cell development. The electrostatic coaction between silver ions (+) and bacterial (-) and explains how nano silver kills bacteria by rupturing the cell wall, releasing the substance of the inner cell, and eventually killing the bacterias that live there [12, 13]. Toxic chemicals used in the production of metallic nanoparticles cause high levels of damage to the environment. According to the green agreement and climate action plan, all polymer-based materials produced must be environmentally soluble and free of toxic content [14, 15]. In this study, silver nanoparticles used to provide an antibacterial effect in filter papers were obtained by green synthesis with plant extracts and raw materials that do not contain toxic chemicals. The ecologically friendly green method for the metallic nanoparticle production is a one-step bio-reduction process. Plant extracts enable bio(green) nanoparticle formation due to their biomolecules including a variety of hydroxyl groups (-OH) (ortho-dihydroxy, hydroxy, carboxy, catechol, amino-) [16, 17]. Terpenoids, flavonoids, phenolic acids, polyphenols, enzymes, amino acids, alkaloids, phenols, carbohydrates, tannins, proteins, or saponins are plant biomolecules that maintain and reduce metal ions (M^+) (M^0) [18]. In the study, the preferred plant extracts for the green synthesis process are Hemp Seeds and St. John's Wort because they are plants with high secondary metabolites. These plant species were selected as a result of previous researches [18, 19]. Each plant species found in nature contains different phenolic groups and organic acids, depending on its variety and type. In the study, Hemp Seeds [20] containing high levels of antioxidants, carboxylic acids, canniprene, and flavonoids and St. John's Wort [21] extracts containing hypericins, flavonoids, hyperforins, antioxidants, mono- and poly-saccharides, phenolic acids were used. It is a proven finding that the pharmacokinetic and therapeutic effects of herbs with different components and secondary metabolites increase more when combined. Despite the fact that this process has not been fully described in the literature, the increased creation of C-O, C-N and C-H bonds with (-OH) groups in various carboxylic and phenolic groups has been identified as a particular instance explanations for the

enhanced impacts of employing plant extracts in combinations [22–24].

The aim of this study is to produce filters used in industrial areas with antibacterial properties by coating of metallic nanoparticles produced using natural ingredients and green production methods. In addition, it has been ensured that this production is safe for human health and natural environmental health, uses low energy and raw materials, and has antibacterial properties. In addition, it is aimed to enhance the mechanical strength of the filter papers by using PVA and nanosilver. Characterization tests, performance tests and antibacterial tests of the produced filters were carried out. Thanks to the determined materials and methods, filter papers with antibacterial effect and high durability that do not harm the environment and human health have been produced.

2 Material and method

PVA has a molecular weight of about 95.000 g/mol and 99% hydrolysis was acquired from Sigma-Aldrich Germany. Silver nitrate (AgNO_3) and all plant extracts are used to synthesize silver nanoparticles. Sigma Aldrich provided the silver nitrate salt (90% concentration). St. John's wort and hemp seeds were purchased from the surrounding market, and all plants were made through DI water wash. Following washing, plants were chopped into tiny bits and dried out for 15 minutes at 80°C in the preheated oven. In order for the acids, metabolites, phenolic groups, minerals and metabolites in plants to be released into water intensively and quickly, the first step to be taken into consideration is to first cut or grind the plants into small pieces. Plants types used in the nano silver and filter fabrication are shown in Figure 1.

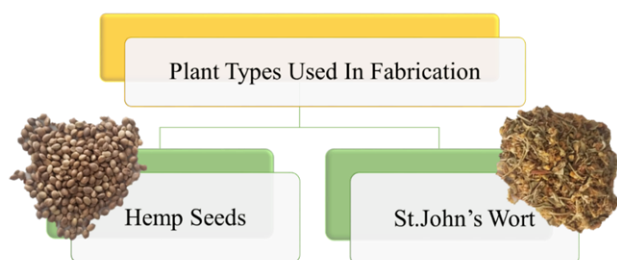


Figure 1. Plants types used in antibacterial filter fabrication

2.1 Green synthesis production of AgNP

In addition to plant extracts, nano silver, one of the most effective metals used to give antibacterial properties to filter papers, was preferred. Nanosilver is produced naturally by the green synthesis method, using only plant extracts, without using any chemicals. While performing nano silver synthesis, the parameters utilized in synthesis used in the previous study were taken into account [18, 19, 25–27]. For the purpose of the creation of plant extracts, 25 g of ground dried herb was inserted to 100 mL of distilled water and kept at 90°C for approximately 45 minutes and after finally filtered. The first procedure in nano silver synthesis is the preparation of 0.1 mM AgNO_3 solution. For this, 0.6 g of AgNO_3 salt should be added to 36 mL of distilled water and

stirred magnetically at room temperature until a homogeneous solution is obtained. 4 mL of plant extract was added to the AgNO_3 solution and continued mixing with a magnetic stirrer. Stirring was continued until the color of the prepared chemical reaction solution changed from pale yellow to dark gray. After 6 minutes, the resulting solution was completely black; this was indicative of the formation of AgNPs. At this point mixing was terminated. The solution underwent filtering and to wash the nanosilver particles ultrasonically, distilled water was used. The final filtered solutions generated nanosilver particles that were dried for two hours at a temperature of 37°C . Figure 2 demonstrates the synthesis procedure of nanosilver particles from plant extracts. The images vividly show the color differences of the solutions. The black particles precipitated at the bottom of the centrifuge tube are green synthesized nanosilvers. Previous studies have proven that nano silvers can be successfully produced by the green synthesis method using plant extracts and their properties [28–30].

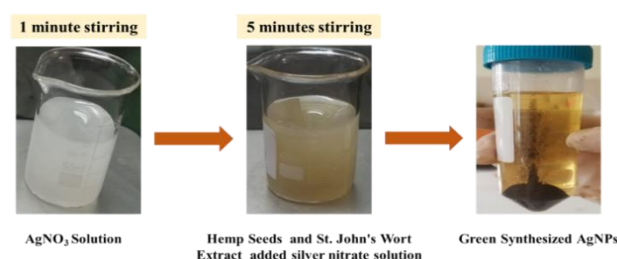


Figure 2. The green synthesis of the nanosilver particles using plant extracts

2.2 Manufacturing PVA/plant extract/AgNPs coating solutions

In the production of antibacterial filters, coating process was carried out by impregnating the coating solutions to the papers by using the solvent casting method. The PVA ratios used in preparing polymer nanocomposite films were prepared with reference from Yontar et.al's [18, 19] studies. Based on comparable researches, the silver nanoparticle adjustment rate employed in the films was identified to be between 2 and 4% [31–36]. These mixing ratios were based on those found in the best quality films. Filter papers are produced by coating with PVA ratios of 2% and 4% in 6 different groups as PVA, PVA and plant extract added and plant extract, PVA and nano silver added. The reason for using 2 different PVA ratios in the study is to investigate the increase the mechanical strength of the papers. It aimed to examine and see how the distribution and amount of PVA in the paper affect the performance values. Sample groups and their contents are given in Table 1 separately by weight. The control sample is commercial filter paper and used for comparisons with other coated sample groups.

Preparing the PVA solution was the initial step in the production process of the coating. To create a homogenous solution, 1 and 2 g of PVA powder were dissolved in 50 mL distilled water in separate beakers at 95°C for an hour. After PVA was dissolved in water, 40 mL of PVA solutions, 10 mL of extracts (hemp seeds and St. John's wort), and 0.25 g of AgNPs were added separately in the beakers.

Table 1. The coated filter paper groups and their contents

Control	PVA-2	PVA-4	PE/PVA-2	PE/PVA-4	PE/PVA/Ag-2	PE/PVA/Ag-4
	1 g PVA	2 g PVA	1 g PVA	2 g PVA	1 g PVA	2 g PVA
Commercial Sample	50 mL DI water	50 mL DI water	40 mL DI water	40 mL DI water	40 mL DI water	40 mL DI water
	-	-	10 mL Plant Extracts	10 mL Plant Extracts	10 mL Plant Extracts	10 mL Plant Extracts
	-	-	-	-	0.25 g AgNPs	0.25 g AgNPs

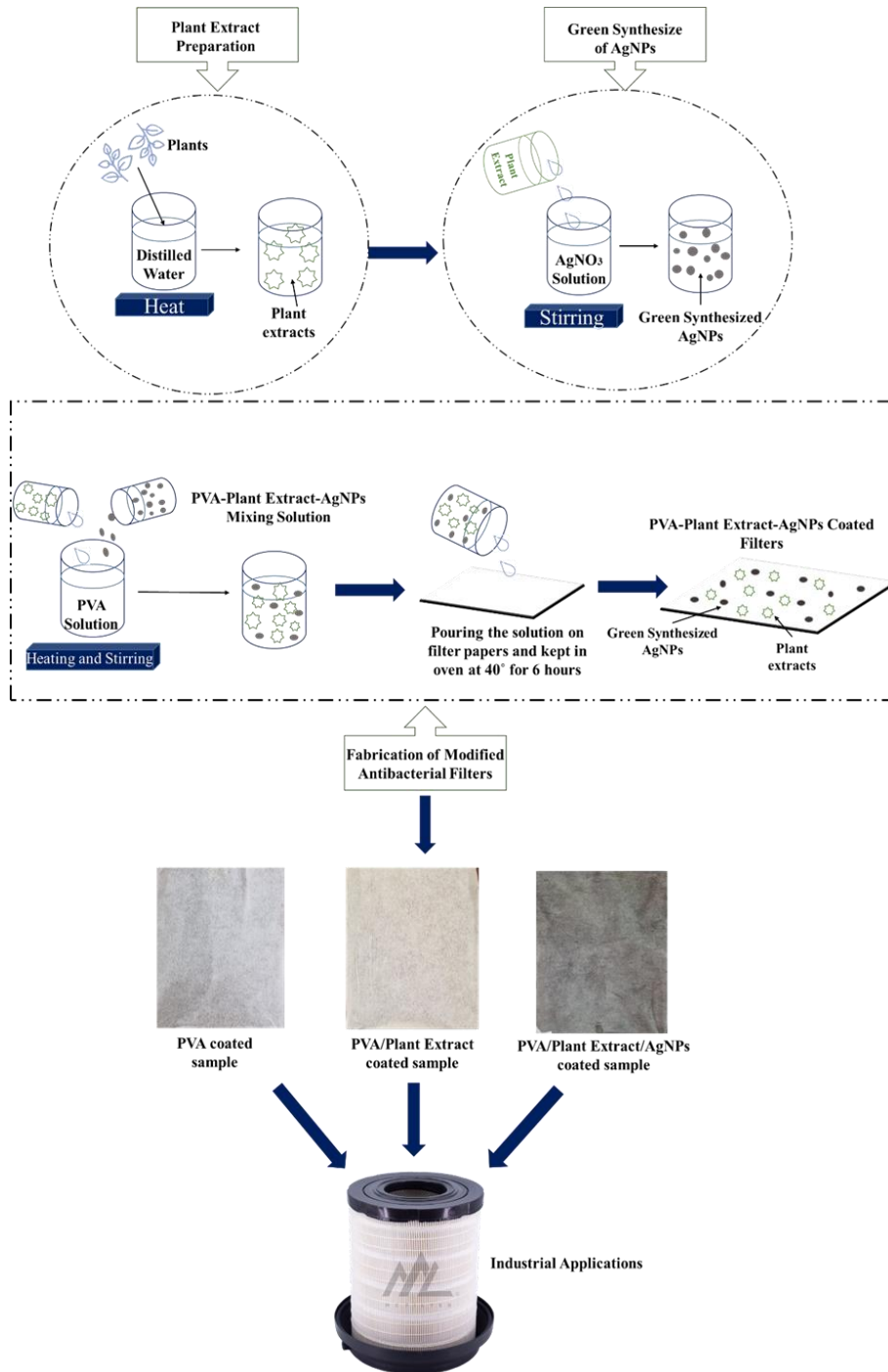


Figure 3. Antibacterial filters fabrication process and produced samples

The resulting mixtures were stirred constantly for 1 hour at 90 °C. Stirring was maintained for 1 hour at 90 °C until all the combined components were homogenous. The polymer coating mixtures were embedded over filter papers, allowed to impregnate for five minutes, and then placed in an oven at 40°C for six hours by hanging the papers vertically in the oven. Figure 3 demonstrates the manufacturing of antibacterial filters and their images.

2.3 Characterizations

A (JEOL 7001F) model scanning electron microscope (SEM) equipped with an EDS analysis adaptor with an 80 mm² X-MAX detector was utilized to evaluate the samples' chemical composition and microstructure incorporating EDS. All of the filter samples underwent SEM examination to determine the size and distribution silver nanoparticles and PVA at all surfaces. Resolution measured under the acceleration voltage of 5 and 10 kV. For imaging in SEM, modified filter papers were divided into 1 x 1 cm squares and coated with gold and palladium. Substances were exposed to chemical analysis using energy dispersive spectroscopy (EDS). EDS spectra and map analysis were used to identify the chemical contents of filter samples and nano silver particles. The presence of Ag elements, plant extracts and PVA on the coating surface of the produced samples was determined by EDS. PVA, nanosilver and plant extracts coatings on the samples were analyzed employing XRD with a Rigaku Smart Lab CuK radiation monochromatic filter to detect their crystal structures at room temperature in the 20°–80° range at a rate of 1°/min and wavelength of 1.2 Å XRD data are collected employing 2D HyPix-3000 area detectors. In order to examine the antibacterial coatings properties developed in the study in XRD, the prepared polymer solutions were turned into thin films by solvent casting method and these films were divided in 1 x 1 cm dimensions. Similarly, these films were cut into 1 x 1 cm dimensions and used for FTIR analysis. The impacts on the structure of silver nanoparticles and extracts of the PVA matrices were evaluated using (Bruker Tensor 27) model Fourier Transform Infrared Spectroscopy (FT-IR) analysis in the 650-4000 cm⁻¹ wavenumber range.

2.4 Performance tests

The performance of the filter papers produced after coating was carried out using devices from M.C.Filter Ltd. company in Samsun/Turkey. All tests were performed 7 times and average values were calculated. In order to determine the permeability properties, the test was carried out on filter paper samples with a diameter of 16 mm and an area of 20 cm² depending on the permeability unit of l/m²/s' under 200 Pa pressure in consensus with the TS ISO 5636-5 standard. The total number of pores and their locations vary according to the modification and coating processes applied to the filters. For this reason, the pore diameter test was applied to the coated filter paper samples. In this test, using Denanol according to TS 4230 EN 24003 standard, the paper was cut in accordance with the chamber diameter of the device and clamped. By adjusting the temperature at a certain rate, the device was stopped at the fixed point. This process

was conducted seven times, with the average measurements of the result gave the pore diameter values of the filters. The mechanical strength of the filter papers coated separately with 2 and 4% PVA ratios can vary with the polymer compared to the control sample. In the high-pressure air cleaning environment, filter papers are damaged due to pressure. As a consequence, it is critical to identify how using PVA in different ratios affects the deformation of the papers under what load. A Burst pressure test was applied to the papers in accordance with TS 3124 EN ISO 2758 standard. The paper is fixed to the device and pressure is applied and the test is terminated at the first tear. The resulting value is in kgf/cm² unit and the maximum load value at which the paper can be used has been calculated.

2.5 Antibacterial efficiency

A modification of the Japanese Industrial Standard JIS Z 2801 was used to test the antibacterial activity of the modified filter papers [37]. The samples were sterilized under a UV lamp for 15 minutes. *E. coli* (ATCC 25922) and *S. aureus* (NCTC-13552) bacteria were prepared at 10⁷ CFU/ml, and 1 x 1 cm test samples were placed in it. A 5 x 10⁶ CFU/ml culture prepared without adding a sample was used as a control. Eppendorf tubes containing samples were shaken at 150 rpm for 4 hours. To separate microorganisms adhering to the sample surface, it was kept in an ultrasonic water bath for 30 seconds and then vortexed. Dilution was made by transferring 100 µL of suspension into 900 µL of physiological saline (FTS). The petri dish containing Plate Count Agar (PCA) was divided into 3 and 25 µL of each dilution was planted, 3 petri dishes were studied. As a result of 24 hours of incubation at 37 °C, the antimicrobial activity value was counted as colonies forming units (CFU). The material without any treatment was taken as a control, and the difference in growth between the polymer-treated culture was evaluated logarithmically. *E. coli* bacteria were planted on TBX agar as a control, and blue colony formation confirmed that it was *E. coli*.

3 Results and discussion

3.1 Characterizations

Modified nanocomposite coating films and control PVA film were divided into 1x1mm squares and investigated by SEM. SEM images of uncoated filter paper were also examined for the purpose of making a comparison with coated filter papers. Figure 4(a) shows SEM images of commercial filter paper. It is also evident from the EDS results of Figure 5(a) that there is no modification on the fibers. It was observed in Figure 4(d-e), the plant extract-derived particles had been distributed uniformly throughout the polymer matrix. The primary C and O components detected in all samples in the EDS spectrum analysis are thought to originate from the chemical composition of PVA and the carbon bands used to fix the samples to the SEM holder. It is also known that they come from the chemical structure of the cellulose used in the production of filter paper. Additionally, it is included in the outcomes of the EDS analysis discovered in related searches [38, 39]. It has been

established from the images of Figure 4(b-c) that the fibers of the PVA-2 and PVA-4 samples are coated with PVA smoothly, only in some regions where PVA is densely deposited at the junction points of the fibers. The fact that the fibers of the PVA-4 sample more condense and frequently than the fibers of the PVA-2 sample is due to the higher amount of PVA used in coating. In PE/PVA-2 and PE/PVA-4 samples, the white particles with different shapes, which are found in large amounts on the fibers, come from plant extracts. In the images of Figure 4(d-e), it is noticed that the plant extract particles have a uniform distribution on both plant extract-modified filter paper surfaces. In addition, Figure 5(d-e) EDS spectrum and mapping results confirm that these particles come from extracts. It is a well-known truth that Hemp Seeds and St. John's Wort plants contain plenty of P, Ca, Mg, K and Fe [20, 21]. The presence of these minerals on the surfaces of these two samples coated with extracts and PVA was clearly demonstrated in the EDS results. While the Pd element seen in the EDS spectrum findings come from the coating made to provide conductivity so that the samples can be viewed in the SEM device. Ti is the contamination product from the external environment to the surface. SEM pictures showed a distinct detection of

AgNPs on the PE/PVA/Ag-2 and PE/PVA/Ag-4 sample surfaces and are shown in Figure 4(f-g). White spherical dots on the filter fibers' surfaces are silver nanoparticles. It is clear from the EDS mapping and spectral results in Figure 5(f-g) that these particles are not just particles from plant extracts. It has been also revealed that silver nanoparticles are present and homogeneously dispersed in the PVA matrix on all two sample surfaces. Silver nanoparticles agglomerated in several areas, however, the amount was minimal degree. Apart from nano silver, the element K, which is captured by EDS spectra comes from plant extracts, and is also present in both samples. SEM and EDS examinations have proven that nanosilver and plant extracts modified with PVA coatings homogeneously dispersed on the surfaces of the fibers. The coating process did not dramatically affect pore closure or distribution. The size and distribution of the pores are critical to successful filtration. The homogeneous dispersion of nano silvers produced by green synthesis on the filter paper as desired is an important step for the emergence of an antibacterial effect. In addition, the presence of the elements from the minerals contained in the extracts on the fibers is important evidence for the green synthesis mechanism and the antibacterial effect.

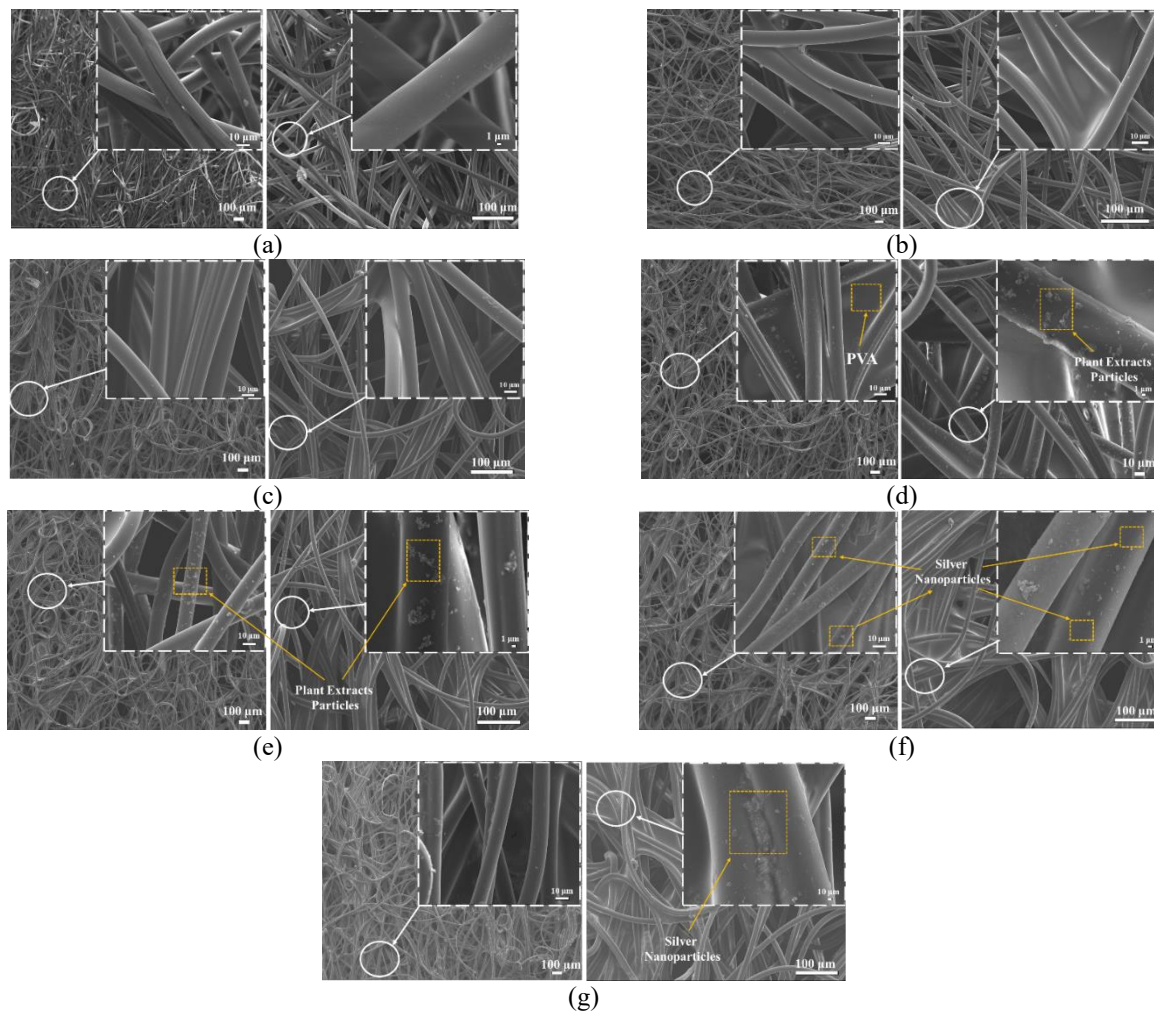


Figure 4. SEM images of (a) Control, (b) PVA-2, (c) PVA-4, (d) PE/PVA-2, (e) PE/PVA-4, (f)PE/PVA/Ag-2 and (g) PE/PVA/Ag-4 filter samples

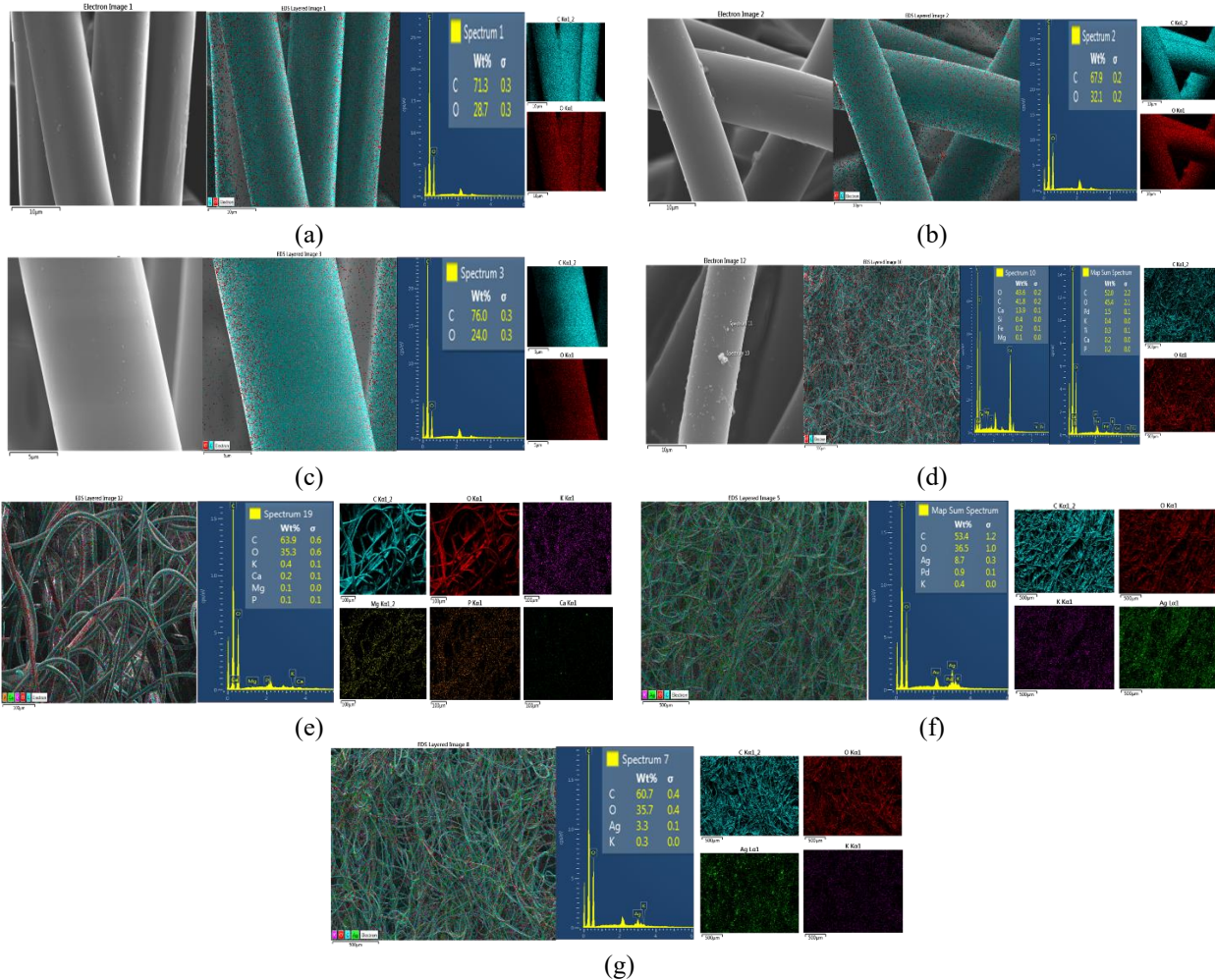


Figure 5. The findings of EDS mapping and spectrum analysis of (a) Control, (b) PVA-2, (c) PVA-4, (d) PE/PVA-2, (e), PE/PVA-4, (f) PE/PVA/Ag-2, (g) PE/PVA/Ag-4 filter samples

XRD analyses were performed on 1x1mm-sized coated filter samples. Characterization tests were selected from the groups with the highest performance characteristics from the coated filter samples. Figure 6 depicts PVA, plant extract-modified PVA, and plant extract and nano silver-modified PVA samples XRD patterns. Regarding the underlying structure of PVA, the high peak reflections at $2\theta = 19.7^\circ$ are derived. These peaks are caused by the crystal planes (101) in PVA's semi-crystal structure. All of the visible peaks at 2θ values of around 38.7° , 45.3° , 63.4° , 76.7° , and 80.9° demonstrate face-centered cubic structure (fcc) of silver nanoparticles. These peaks resemble Bragg's reflections (311), (220), (200), and (111), respectively. Peak broadening in the X-ray diffraction spectrum is frequently related to smaller particle sizes. Silver nanoparticles in nanocomposite coated samples are readily apparent in the XRD patterns. Because crystal structure formation did not occur, sharp peaks were not created in the XRD results for the PE/PVA-4 sample.

All 1x1mm square samples were evaluated and interpreted using FTIR analysis. The transmittance band at 3253 cm^{-1} in the FTIR spectrum (Figure 7) is equivalent to the OH stretching vibration generated by pure PVA. The

peaks at 2929 cm^{-1} and 2905 cm^{-1} are equivalent to symmetric and asymmetric stretching of the C-H alkyl group, respectively. Likewise in the neat polymer structure, the transmittance peak at 1637 cm^{-1} is induced by the stretching vibration of the acetate C=O group. The 1443 cm^{-1} peak is due to CH_2 bending, the $955\text{-}1160\text{ cm}^{-1}$ band is a result PVA acetyl C-O stretching, and the 833 cm^{-1} transmittance peak is due to the original polymer's rocking C-H band [40]. The heterocyclic compounds' C=C bonds' stretching vibrations in St. John's Wort-Hemp Seeds extracts caused the formation of a new adequate band at 1644 cm^{-1} in the case of PE/PVA-4 and PE/PVA/Ag-4 films. The detected rise in the peak of absorption of C=C bonds may be attributed to the functional groups found in plant extracts. The plant extracts decreased the stretching vibration bands of the C-O and O-H bonds in the coating material structure, displaying that the extracts influenced the PVA structure. The new bonds formed by the HO- bonds of secondary metabolites derived from the PVA molecular chain and plant extracts ended up in C=C bonds rather than C=O vibrations [19]. The appearance of hyperforin, carbohydrates, phenols, alcohols, or water molecules from plant extracts results in the existence of a wideband associated with OH group stretching

vibrations that exist between 3450-3250 cm^{-1} [41, 42]. Following the AgNPs' immediate integration into the PVA matrix, certain polymer chain bonds ruptured, and new chains were produced in the vibrations of the C-H, C=O, and O-H groups in the PVA structure. When comparing the spectra of AgNPs modified PE/PVA/Ag-4 samples, the strength of the vibrational bands at 2941, 1722, 1424, 1322, 1038, and 833 cm^{-1} , which are equivalent to the stretching vibration of CH_2 , is clearly visible. As shown by the C=O stretch, C-H breakage vibration, C-H bending vibration of CH_2 , C-C stretching vibration and the C- O stretching of acetyl groups of PVA, the molecules of the polymer were successfully adsorbed onto the outermost layer of the silver nanoparticles [18, 43]. The rise in the FTIR spectrum between 1350 and 1650 cm^{-1} is equivalent to the CH_2 and C-H bond and shows fractured chains. In the PE/PVA/Ag-4 sample, AgNPs created new bonds in this range. Since oxygen atoms offer duplet filling in an empty space on the outer layer of silver atoms as well as benzene rings are present in plant compounds, the band of C=O weakens and moves from 1712 cm^{-1} to 1649 cm^{-1} . Plant extract molecules and nanosilver disrupted the PVA chains and established new bonds, according to all FT-IR spectra.

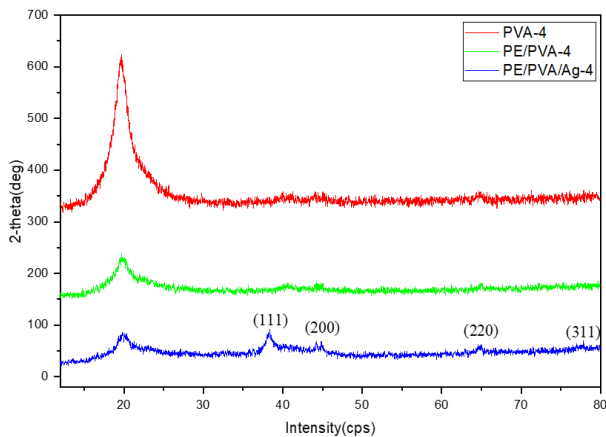


Figure 6. XRD results of neat PVA film and antibacterial filter coating films

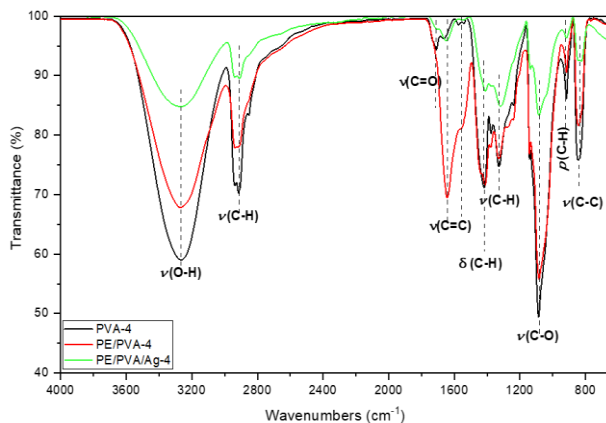


Figure 7. FTIR results of neat PVA film and antibacterial filter coating films

3.2 Performance tests

The high air permeability of the filter papers used in the industrial industry and automotive provides an increase in the amount of cleaner air and a faster filtration process. The effects of PVA and nano silvers, which were used in the study to provide an antibacterial effect and high mechanical strength, on the permeability properties of the filters are shown in the graph in Figure 8(a). The filters with the highest permeability compared to the control sample were PVA-4 and PE/PVA/Ag-2. The average pore diameters of the PVA-4 and PE/PVA/Ag-2 samples were calculated to be only 3% lower than the commercial standards of the filter paper. This can be explained by the coating of PVA and nano silvers on the fibers, providing sufficient porosity without filling the filter paper pores. Although it was thought that PVA would reduce the permeability by filling the voids of the fibers, on the contrary, the porosity values were increased with the addition of high adhesiveness and nano silver, resulting in a low decrease in the amount of permeability. Although this decrease is an expected result, it is higher than the results obtained in similar studies [44, 45] and with the applied coating only 11% and 13% reductions occurred in the permeability amounts of these samples, respectively. The fact that the pore sizes of filter samples coated only with PVA are lower than those of nano silver modified samples is due to PVA filling the gaps between the fibers by forming a film. Nano silver particles disperse in the PVA structure and form chemical bonding. With binding, the amount of water molecules in the PVA chains decreases. Therefore, a more compact structure is created. This enables PVA modified with nano silver to be coated on the fiber surfaces rather than filling the gaps between the fibers. Thus, the higher amount of pores causes the permeability rate of samples containing nano silver to be higher.

It is clear from the graph in Figure 8(b) that there is no direct proportionality between permeability and pore diameters. The homogeneity of the coatings is another parameter that affects these values. However, it was determined that there was a very low decrease in pore diameters with coating applications. Compared to the control sample, the sample with the smallest pore diameter is PVA-4, while the sample with the highest pore diameter is the PE/PVA/Ag-4 sample. It has been understood that the smaller or higher pore diameter does not adversely affect the permeability. As can be seen in the SEM photographs shown in Figure 4, it was determined that PVA filled the fiber gaps in some regions and this caused a decrease in the pore diameter. The pore diameters of the PE/PVA/Ag-2 and PE/PVA/Ag-4 samples were very close to the control sample values, thanks to the porosity of the plant extracts and silver particles with high surface areas and nano-sizes, and the homogeneous coating on the surfaces by wetting the fibers better. As a result of burst pressure tests of the coated filter papers, it was determined that all coated samples had higher strength than the control sample. As seen in Figure 8(c), PE/PVA/Ag-2 sample had the highest burst strength among the coated samples.

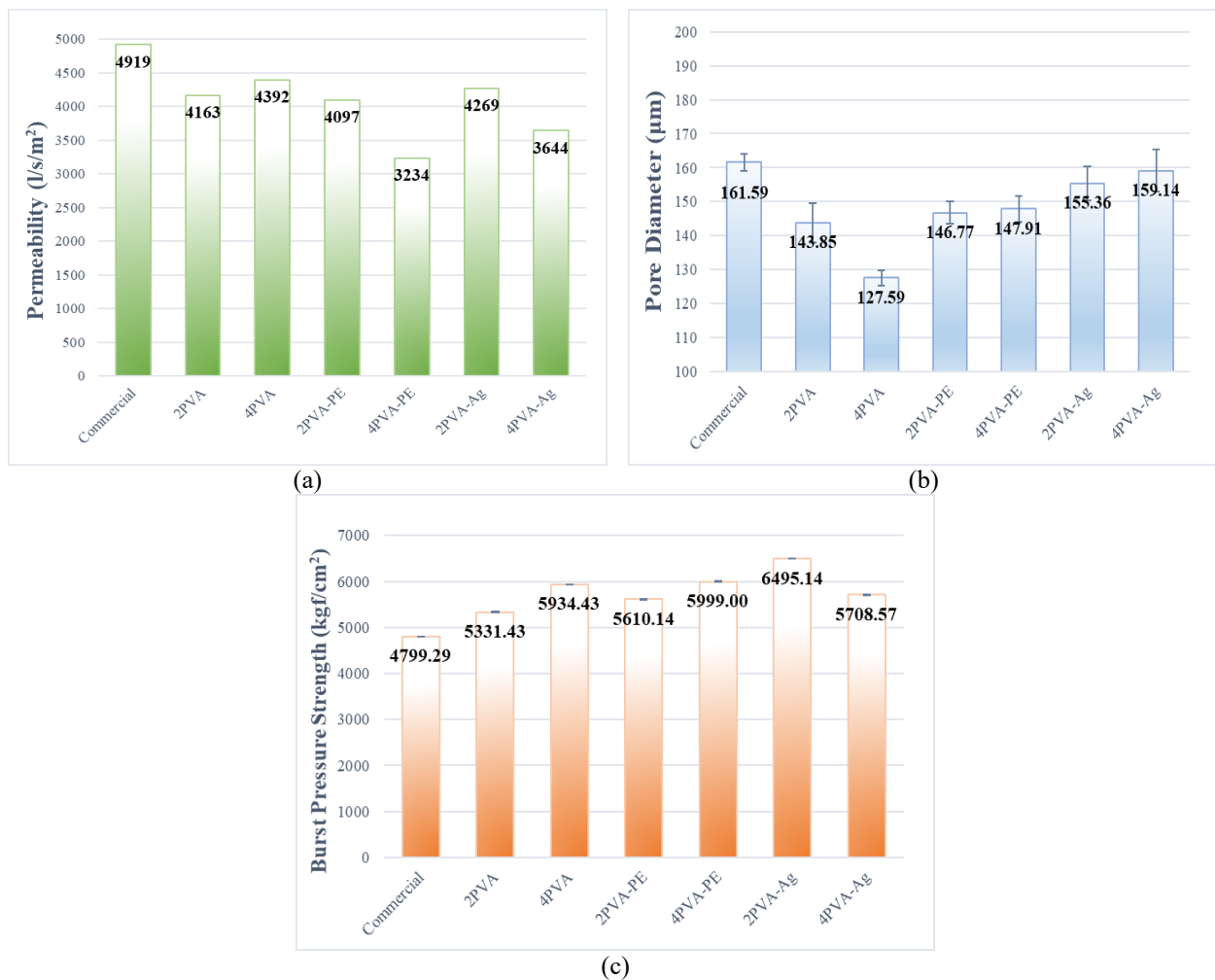


Figure 8. Performance test results, (a) permeability, (b) pore diameter and (c) burst pressure strength

Silver nanoparticles and plant extracts bonded with PVA to form a strong polymer structure and increased the resistance of the filter paper against pressure with the homogeneous distribution of this structure on the fibers. As it is known, nanocomposites prepared with nanoparticles and plant extracts have higher mechanical strengths than unmodified ones. The reason for this can also be shown to be the cross-linking of nano-sized particles with polymer chains [13, 18, 19, 46, 47]. In this way, the strength of the coated filter papers has also increased. Performance test results of all samples revealed that PE/PVA/Ag-2 had 36% higher burst strength than the control sample and was the sample with the closest permeability and pore diameter to the control sample on average.

3.3 Antibacterial efficiency

Figure 9 depicts photos of bacteria growth on agar conducting antibacterial evaluation. Six unique bacterial counts were implanted in the medium of each sample. These bacteria are represented by the numbers $\times 10^6$, 10^5 , 10^4 , 10^3 , 10^2 , and 10^1 . After the antibacterial test, the specimens concentrations and logarithmically decreased numbers of bacteria in the medium are shown in Table 2. Furthermore these decrease rates are visually depicted in

Figure 10. It was determined that the PE/PVA/Ag-4 filter paper sample containing silver nanoparticles had the highest Log_{10} reduction. It inhibited bacterial growth at a value of Log_{10} as 4.04 (10,000 times) against *E. coli* and 3.17 (1000 times) against *S. aureus*. The significant antibacterial activity is likely ascribed to the AgNPs on the filter surface, which can effectively suppress the proliferation of bacteria by damaging the membrane of bacterial cells [48]. Previous research has indicated that antibacterial products will have different antibacterial activity in the presence of *S. aureus* and *E. coli*. Because *S. aureus* contains a stronger cell wall and reduced protein content, it is more difficult to attack by silver ions [49] Hence, the silver-containing filter paper has lower Log_{10} values for *S. aureus* than for *E. coli*. The PE/PVA-4 sample, which does not contain nano silver, was coated only with plant extracts and PVA. The growth reduction ratios of PE/PVA-4 sample against *E. coli* and *S. aureus* were 1.26 and 2.14. This finding demonstrates that the St. John's Wort and Hemp seeds extracts had been quite effective to give antibacterial properties to the filters. The reason why the uncoated commercial filter paper seen in Figure 9(b) also has a low antibacterial effect may be due to its contact with other test samples.

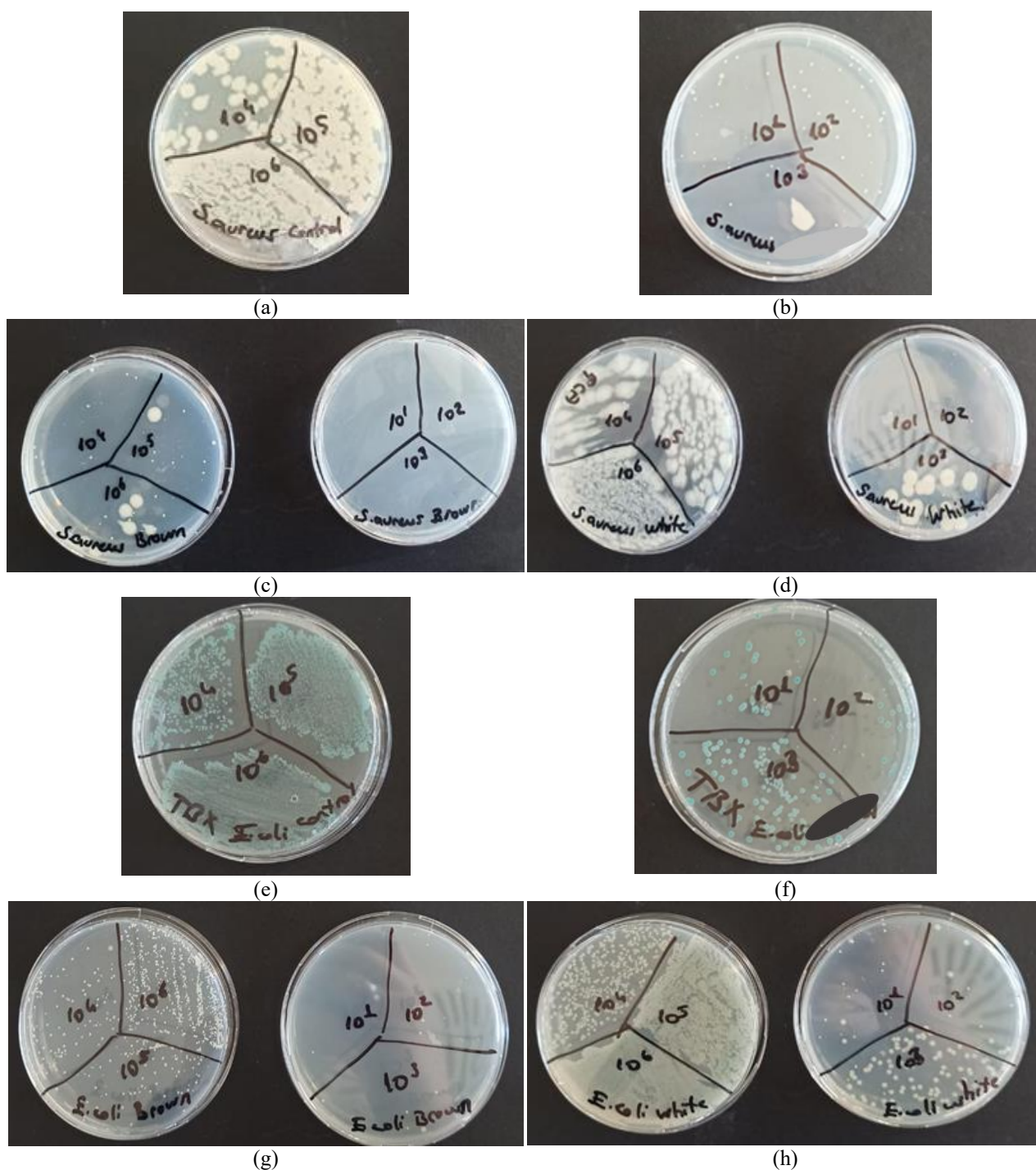


Figure 9. Antibacterial activity tests of (a) control *S.aureus*, (b) uncoated commercial filter paper against *S.aureus* (c) PE/PVA/Ag-4 against *S.Aureus*, (d) PE/PVA-4 against *S.aureus* , (e) control *E.coli* , (f) uncoated commercial filter paper against *E.coli* , (g) PE/PVA/Ag-4 against *E.coli*, and (h) PE/PVA-4 against *E.Coli*

Table 2. Inhibition of *S. aureus*(S) and *E. coli*(E) by log CFU/mL of plant extracts and AgNPs modified filter samples for first antibacterial test method

Bacteria	Control	PE/PVA/Ag-4 Sample Concentration (CFU. ml ⁻¹)	Log ₁₀ reduction	PE/PVA-4 Sample Concentration (CFU. ml ⁻¹)	Log ₁₀ reduction
<i>E. coli</i>	4.4±0.2x10 ⁷	4.0±0.2x10 ³	4.04	2.4±0.4x10 ⁶	1.26
<i>S. aureus</i>	4.2±0.4x10 ⁷	2.8±0.2x10 ⁴	3.17	3.0±0.2x10 ⁵	2.14

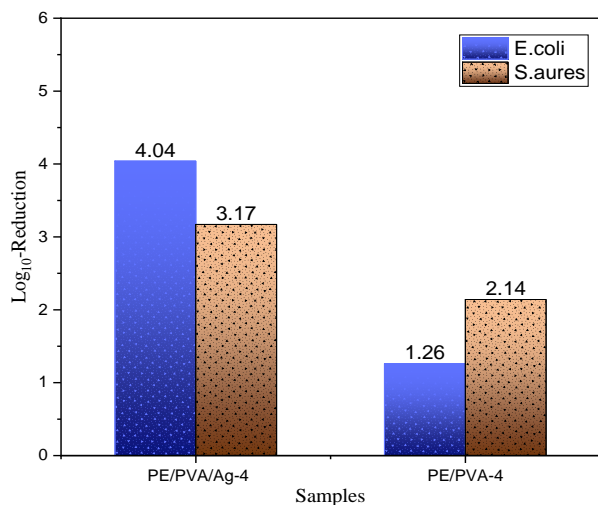


Figure 20. Log CFU/mL of coated filters against *E.coli*(E). And *S.aureus*(S) bacterias

4 Conclusions and suggestions

The objective of this research was to develop green, nature-based, eco-friendly, and antimicrobial fiber filters. The bio-synthesized silver nano particles were spherical in form and uniformly scattered on the fiber surfaces, according to SEM and EDS examination. FTIR outcomes also showed that green synthesized nano silvers and plant extracts broke the PVA chains and formed new bonds, and as a result, a successful coating material was produced. The existence of nano silvers generated by biosynthesis was proved by also XRD analysis. With the coating process, particles derived from plant extracts and nano silvers were homogeneously distributed on the fibers. As expected, modification of nano-silver and plant extracts has been shown to impart antibacterial properties to the filters. In addition, a high antibacterial effect was achieved with the coatings made using only plant extracts. In this way, the production of highly effective antibacterial filter papers that are harmless to the environment will be paved with the use of plant extracts. The performance tests of filter papers coated with plant extracts, nano silver and PVA were also higher than the other samples. The most important of these features is burst pressure resistance. Thus, while the service life of the filter papers will be increased, it will also provide antibacterial effects. As a result, it has been demonstrated that environmentally friendly nano silvers and plant extracts may be used to create antibacterial paper filters for industrial applications without the need of any hazardous chemicals.

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Conflict of interest

The authors declare that there is no conflict of interest

Similarity rate (iThenticate): 8%

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