**Bio-adhesive coating from Ethylene Propylene Diene terpolymer (EPDM)/Expanded Organoclay to Polyester Fabric for Multi-Applications**

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**Abstract**

Two methods were used for preparation of bio-adhesive coating from Ethylene Propylene Diene terpolymer (EPDM)/Organoclay nanocomposites rubber mix. First method includes modification of the nonpolarity of EPDM by using polar Ethylene Propylene Diene terpolymer grafted Maliec Anhydrite EPDM-g-MA and another one includes green modification of organoclay by stearic acid Expanded Organoclay (EOC). These mixes were loaded by long bio-alkyd resin based on soyabean oil (LAR), dry bonding system consisting of hexamethylene tetramine, resorcinol, hydrated silica (HRH). The mixes were charged by different amounts of organoclay (OC) or expanded organoclay (EOC) according to each method. The prepared rubber nanocomposites mixes were then spread over polyester fabric, cured forming of the final proofed product. The peel strength was used to measure the adhesion properties between the EPDM rubber and polyester fabric. Other measurements such as barrier properties, antimicrobial and flammability were conducted on the end proofed fabrics. The obtained results showed that the addition of soyabean long bio-alkyd resin, HRH bonding system and organoclay improved the adhesion strength between the EPDM rubber and the polyester fabric. The final proofed fabric could strongly be a candidate for various applications.

1. Introduction

Adhesion of rubber compounds to reinforcing fabric, is one of the most important technique in rubber industry [1]. The rubber fabric adhesion is a hydrogen bond, Vander Waaals forces and crosslink bonds [2,3]. Better rubber-fabric adhesion can be achieved by using the highest bonding system and the highest adhesion promotor. The rolle of the adhesion promotor is to increase the wetting of substrates in addition of chemical bonds between rubber and fabric substrates [4]. The authors studied before [5-8] the use of various oil resions as promotors. These are natural biodegradable resins; named soyabean oil, sun flower oil, olive oil and coconut oil. The selection of these oils was due to their green economic value, low cost, wide availability, and eco-friendly behaviours [9-14]. According to their viscosity and chain, the resins were classified to short, medium and long oils. The study showed that using long oil alkyd resin (soyabean resin) as promotor, the rubber fabric achieved better adhesion properties. In spite of these results, there was some drawback properties such as high gas permieability, low thermal stability, high flammability and slit UV degradation. To avoid their drawback, the authors in this research used nanofiller to overcome these disadvantigies. Organoclay (OC) was used in different concentrations, fixed amount of long bio-alkyd resin as reinforcing substrates. The use of OC as nanofiller can act much better than using unmodified layer silicate (i.e montmorillonite, MMT). It was repoted before the OC is cmpatable with the hydrophobic EPDM elastomer. Morphological studies showed also that EPDM/OC nanocomposites showed two different interaction phases, one is phase separated and the other is exfoliated structure [17]. The major advantages in using nanocomposite are the lighter weight of the final product. This is due to the use of low filler loading of nanoparticles and the large surface area of the nanoparticles [18]. For better interaction between EPDM and OC, the polymer must be grafted by maleic anhydride to form EPDM-g-MAH [19]. There is another method by using non-toxic intercalate agent (e.g stearic acid). This is to expand the interlayer spacing distance of OC and to form expanded OC (EOC). This method modifies the nonpolar EPDM or facilitate the rubber chains to peneterate EOC layers, leading to intercalated/or exfoliated structures. This by turn increases the rubber-filler interaction. The expansion process of OC layers can be occurred by two pathways: i) by increase the content of long chain alkyl ammonium salts, but this causes several issues such as at high content of organomodifier the presence of Br-1 toxic anion between clay layers which leads to limited use in the industrial applications [20]. The decomposition of alkyl ammomnium salt at temperature 170-180 º C which cause unaccepted color and odour, which considered as another problem in the industrial applications [22,23]. ii) By further green modification of organoclay using of stearic acid as nontoxic surface modifier which facilate the intercalation of EPDM rubber into organoclay gallery. Hydrophobic tail of stearic acid is compatible to hydrophobic EPDM rubber, by turn better interaction between EPDM and modified organclay [17]. The use of HRH bonding system as dry additive on rubber mix is highly preferable than chemical dipping treatment of fabric for improvement the adhesion strength of rubber coated fabric [18]. The effect of dry three component systems, composed of resorcinol, hexamethylenetetramine and hydrated silica on the adhesion of NBR to nylon cord was studied by Darwish et al [24]. Other pervious researches have reported the inferior adhesion strength between EPDM and polyester fabric [25]. However, HRH internal bonding system, LAR adhesion promotor and OC of different concentrations were used with other ingredients. The mix was carried out on two-roll mill. These mixes were immersed in equal weight of toluene to form the rubber dough. The dough was spread over the polyester fabric, cured to form the proofed fabric. The final product will be subjected to different mechanical, physical and chemical measurements. The proofed fabric material has many applications in industry; it can be used in building sectors as roofing membranes, transport sector, hospitals, and electrical engineering insulations.

**2.** **Experimental**

2.1. Materials and preparation methods

Ethylene propylene diene rubber(EPDM) was composed of 53 ± 4 wt% ethylene content and 6.5 ± 1.1 wt% ENB content with ethylidene norbornene as termonomer. This polymer was supplied by LANXESS Deutschland Gmbh, Germany (BUNA®EP T9650). Maleic anhydride grafted ethylene propylene diene terpolymer (EPDM-g-MAH) was supplied from Uniroyal Co. under Royaltuf's reference (498) and purified as described in our previous literature [19]. The adhesion promoter used is soya bean long oil alkyd resin (LAR). This resin was supplied by EGALE CHEMICALS™, Egypt. The organoclay used is (Nanoclay, surface modified contains 25-30 wt.% methyl dihydroxyethyl hydrogenated tallow ammonium). This nanoclay was supplied by Sigma-Aldrich, Egypt. Hexamethylenetetramine (HMT) of purity 99% was purchased from Alfa Aesar, Gremany. Stearic acid which used as an expanded toxicity-free for organoclay was purchased from Sigma-Aldrich, Egypt. All other reagents were provided by El Nasr Pharmaceutical Chemicals Co., Egypt. The polyester fabric used was supplied by Misr Helwan for textile, Egypt.

**2.1.1. Preparation of Expanded Organoclay (EOC)**

To increase the interlayer spacing distance (d-spacing) of organoclay (OC), stearic acid was mixed with OC in a porcelain mortar using a mortar pestle as described in our previous literature [20]. Different contents of organoclay (i.e. 3, 5, 7, and 10 wt. %) were mixed with the stearic acid (SA) in the ratio of 1:1 to obtain toxicity-free expanded OC. The blend was heated in the oven at 110ºC for 60 min. Then, the blend was continuously stirred using a pestle in the oven. To obtain much homogeneously mixture, the process of heating and stirring was repeated three times to ensure that melted stearic acid (SA) intercalated into the gallery of organoclay. After cooling the expanded organoclay (EOC) crushed in an electric grinder for about 3 min at the room temperature. The obtained EOC is characterized by using XRD and FTIR techniques.

2.1.2.  **Preparation of nanocomposites samples**

Two methods were used to prepare the EPDM /Alkyd resin / Organoclay nanocomposites. The first method is known by EPDM/EP-g-MAH/ OC/LAR (called P/EPg/OCx). In this method, after the premastication of EPDM rubber using water cooled two-roll mill (152.4 x 330.2 mm) at a gear ratio 1:1.4., the 10 wt. % of EPDM-g-MAH was added. This was followed by additing 20 wt.% of LAR resin for all mixes. The mix formulations are given in Table 1. Other ingredients were orderly added according to Table1. In the second method, the same mix formulation that given in Table 1 was used, except the EPDM-g-MAH and OC are replaced by EOC modified filler. The samples were abbrievated as P/EOCx, where x refers to the filler amount (i.e 3, 5, 7, and 10 wt.%) in the mix, as shown in Table1.

**Table 1.** Mix formulations of EPDM/EPDM-g-MA/OC/LAR and EPDM/EOC/LAR nanocomposites.

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **P/EOC10** | **P/EOC7** | **P/EOC5** | **P/EOC3** | **P/EPg/**  **OC10** | **P/EPg/**  **OC7** | **P/EPg/**  **OC5** | **P/EPg/**  **OC3** | **Unfilled EPDM** | **Ingredients**  **(wt. %)** |
| 100 | 100 | 100 | 100 | 90 | 90 | 90 | 90 | 100 | EPDM |
| - | - | - | - | 10 | 10 | 10 | 10 | - | EPDM-g-MAH |
| 10 | 7 | 5 | 3 | - | - | - | - | - | EOC |
| - | - | - | - | 10 | 7 | 5 | 3 | - | OC |
| 20 | 20 | 20 | 20 | 20 | 20 | 20 | 20 | 20 | LAR a |
| 1 | 1 | 1 | 1 | 1 | 1 | 1 | 1 | 1 | Stearic acid |
| 5 | 5 | 5 | 5 | 5 | 5 | 5 | 5 | 5 | Zinc oxide |
| 5 | 5 | 5 | 5 | 5 | 5 | 5 | 5 | 5 | Resorcinol |
| 3.2 | 3.2 | 3.2 | 3.2 | 3.2 | 3.2 | 3.2 | 3.2 | 3.2 | HMT b |
| 5 | 5 | 5 | 5 | 5 | 5 | 5 | 5 | 5 | Hi-Silica |
| 0.75 | 0.75 | 0.75 | 0.75 | 0.75 | 0.75 | 0.75 | 0.75 | 0.75 | MBT c |
| 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | TMTD d |
| 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | Sulfur |
| **Rheometric behavior at 160ºC** | | | | | | | | | |
| 4:56 | 6:10 | 5:10 | 4:40 | 9:07 | 9:25 | 8:22 | 8:34 | 6 :55 | Cure time  T90, min:sec |
| 10.31 | 9.03 | 9.47 | 9.36 | 8.60 | 8.01 | 8.38 | 7.43 | 7.52 | Maximum torque |
| 0.36 | 0.44 | 0.50 | 0.60 | 0.72 | 0.63 | 0.50 | 0.68 | 0.93 | Minimum torque |
| 0:45 | 0:50 | 0:48 | 0:43 | 1:00 | 1:03 | 1.00 | 1:00 | 0:57 | Scorch time, ts2 min: sec |

a Long oil alkyd resin based on soya bean oil, b Hexamethylene tetramine,

c 2-Mercapto benzo thiazole, d Tetramethylethiuram disulfide

**2.1.3. Rubber dough preparation**

Equal weight of unvulcanized rubber mix and toluene were mixed together to prepare the rubber dough. After 24-48 hours, the rubber dough was formed and stirred manually to increase the dough homogeneity.

**2.1.4. Spreading technique**

The spreading machine was used for the preparation of rubber coated fabric. The rubber dough was spread over the polyester fabric. The fabric thickness in 0.25 mm. By adjusting the two vernires, a thin rubber layer of 0.3 ± 0.1mm was spread over the warp direction of polyester fabric. The rubber coated fabric was folded to form fabric-rubber- rubber-fabric sheet (thickness 0.8mm ± 0.2mm).

**2.1.5. Vulcanization process**

The rubber proofed fabric was wrapped with cotton linen cloth and rolled around a metal drum then blanketed again with cotton to prevent warping during the curing process. The drum was then transferred to an air-circulation oven adjusted at the optimium temperature of 160ºC and for the duration of time estimed from the Monsanto Rheometer (MDR 2000) results that supplied by Alpha Technologies, UK, in accordance with ASTM D2084-07 standard at 160°C, as reported in Table1.

**2.2. Characterization techniques**

**2.2.1. X-ray diffraction (XRD)**

The XRD measurments were conducted using a Philips X-ray diffractometer PW 1930 generator, PW 1820 goniometer, equipped with Cu Kα radiation (45 KV, 40 mA, with λ=0.15418 nm). The XRD analysis was used to detect the d-spacing distance and 2Ɵ position of OC and EOC prepared and how far the interlayer space of OC was affected by the stearic acid. The Scan of the analysis was run in the 2θ range of 2.5 to 10º with step time of 2s.

**2.2.2. Fourier transform infrared spectroscopy (FTIR)**

The FTIR-ATR Analyzer, model Cary 630 FTIR spectrometer, Agilent Technologies Company, was used to analyze the prepared samples at the IR scale. The spectral range was from 4000 to 550 cm-1 with a resulation of 4 cm-1 and 32 scans.

**2.2.3. Adhesion strength measurements**

The Peel test at 180º was used to measure the adhesion bond of EPDM rubber to polyester fabric sheet. Strip samples of 250 mm length and 25 mm width were cut from the rubber coated fabric. The separated end of the test sample was clamped into position on Zwick (Model Z010) Tensile Testing Machine, Germany and pulled at a speed rate of 50mm min-1 at a temperature of 23 ± 2°C. The adhesion peel strength was expressed as N 25mm-1. The adhesion strength measurements were performed according to ASTM D413. From one sheet, the average peel strength of five measurements for each sample was recorded.

**2.2.4. Scanning electron microscopy (SEM)**

The morphology of the peeled surface for rubber-coated fabric samples was investigated using scanning electron microscopy SEM-FEI Model Quanta 250 FEG (Field Emission Gun), Netherlands. The accelerating voltage of 30 kV and spot size (3-3.5) were carefuly chosen to optimize the quality of pictures. The surfaces of the investigated specimens were examined without coated gold and at low vacuum.

**2.2.4. UL 94 measurments**

The flammability performance for rubber-coated fabric samples was carried out using UL94 flame chamber (Stanton Rheometric Scientific, LTD, UK) in a horizontal position in accordance with ASTM D 635-14. The strip rubber-coated fabric size was 150 x 50 x 0.25 mm3. The rate of burning was recorded for at least five samples.

**2.2.5. Antimicrobial activity test**

To assess the antimicrobial activity of fungal extracts, the Disc agar plate method was used [26]. The rubber coated fabric strip (unfilled EPDM, EPDM -g-MA/ LAR/OC5, EPDM /LAR/ EOC5) of diameter 0.5cm was used in the antimicrobial activity test. The rubber coated fabric samples were examined against three different microbial strains, i.e., Gram-positive bacteria (Staphylococcus aureus, G+ve bacteria), Gram-negative bacteria (Pseudomonas aeruginosa, G-ve bacteria) and fungi (Candida albicans). Both bacteria and fungi test microbes were grown on nutrient agar (DSNZ 1) medium (g/L): beef extract (3), peptone (10), and agar (20). Whereas fungal test microbe was grown on Szapek-Dox (DSMZ130) medium (g/L): sucrose (30), NaNO3 (3), MgSO4.7H2O (0.5), KCl (0.5), FeSO4 7H2O (0.001), K2HPO4 (1) and agar (20). The culture of each microorganism was diluted by sterile distilled water from 104 to 108 CFU/ml to be used as inoculum. 0.1ml of the previous inoculum was used to inoculate 1L of agar medium (just before solidification) then poured in Petridishes (10 cm diameter containing 25ml). Discs (5 mm diameter) were located on the surface of the agar plates previously inoculated with the test microbe and incubated for 24 h for bacteria and fungi at 37 °C.

## Result and discussion

### X-ray diffraction (XRD) analysis

XRD technique was used to ascretin the intercalation of stearic acid (SA) between the interlayer spacing of organoclay layers. Fig. 1 shows the shift of the basal spacing (d-spacing) of EOC specimen that can confirm the inserstion of SA particles into clay galleries. The organoclay (OC) has a d-spacing of 1.8 nm at around 2Ɵ = 4.9º before its modifying with SA. After the modification, a large increment in the d-spacing distance to 4 nm at 2Ɵ = 2.2º is noticed, evidencing that the SA chains are obviously intercalated within OC to expand d-spacing distance of OC, as shown in Fig. 1 [20]. This renders that the non-polar EPDM chains are more compatible with EOC and facilitates their interactions to polyester fabric.

**Figure 1**. XRD patterns for OC and its expanded EOC (Ref. 20)

* 1. **FT-IR analysis**

The functional groups of expanded OC, LAR resin, polyester fabric, and their proofed fabric samples, as well as the chemical interaction between them were studied by ATR- FTIR spectra within the range from 4000 to 600 cm-1, as indicated in Fig. 2. From the figure, it is noticed that OC and EOC have the same absorption bands at 3640 cm-1 for crystal OH, at 3420 cm-1 for OH stretching of organomodifier, and a massive peak appearing at 1051 cm-1 corresponding to Si-O-Si group [19,20]. However, a new peak appears at 1715 cm-1 which assigned to C=O group for stearic acid [20]. This confirms that strearic acid molecules intercalated in the clay galleries. The figure also shows the spectra that are corresponding to bio-alkyd resin (LAR), in which the main bands appear at 3457 cm-1 for free OH, at 1736 cm-1 for C=O, and at 1606 cm-1 for C=C of unsaturated fatty acid. Regarding to the intensity of C=O in the proofed fabric samples (i.e. P/EPg/OC5 and P/EOC5), it is reduced with addding of the nanofiller. On the other hand, the OH and C=C bands in LAR and polyester are disappeared. This implies that good chemical interactions between the functional sites of rubber/nanofiller/LAR/fabric are occurred , leading to a great enhancment in the adhesion properties. These results are consistent with that obtained by peel properties.

**Figure 2**. FT-IR spectra for proofed EPDM/polyester fabric based on LAR and nanofiller systems.

**3.3. Adhesion properties**

**3.3.1 Effect of nanofillers on peel properties**

It was reported before [27] that applying 20 % by weight of MT black as filler, is an optimum loading for adhesion. However in this study, 5 % by weight of EOC or OC compounds showed better adhesion properties. This can be shown in Fig. 3. The reason is that, the presence of expanded OC or OC compounds, acts as reinforcing and compatibilizing agent in the mix. In addition to that, the stearic acid or the organomodifier in the clay galleries, act as terminal group between the bio-alkyd resin and the polyester fabric. This adds new chemical bonds to the matrix and by turn higher adhesion properties, which confirmed by FT-IR technique. Fig. 4 shows different adhesion peel strength values for two different systems EOC and EPg/OC, where the former exhibits higher peel strength than the later. It is likely significant that the two systems recorded maximum peel strength at 5 wt.%, with higher peel strength for P/EOCx system.

**Figure 3.** Peel force as a function of peel strain for proofed samples (P/CB20, P/EPg/OC5 and P/EOC5).

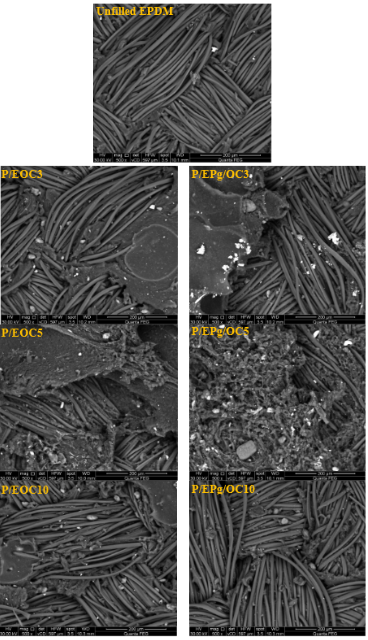
**Figure 4**. Peel strength as a function of filler content for two proofed fabric regimes.

**3.4. SEM analysis after peel tests**

SEM can reveal interesting information about the morphology of proofed fabric specimens and embedding degree of the filled EPDM by EOC or OC at different filler loadings within the polyester fabric. SEM micrographs representing the investigated specimens are shown in Fig. 5. The morphology of unfilled proofed fabric specimen displays less adhered EPDM elastomer between the filaments of polyester fabric, indicating poor adhesion at phase boundaries. This is an indication of the inmiscibility and incompatibility of the polymer within the filaments of the fabric in the absence of expanded OC [19]. With introduction of 3 wt.% of OC or EOC to the matrix, a partial wettability of elastomer onto the filaments is observed for the both regimes. When the amount of OC or EOC in the matrix increased to 5 wt.%, EPDM rubber seems to be embedded and more compatible with the polyester fabric, showing strong adhesion between them. This indicates that the green modification of OC by stearic acid or adding EPg/OC is more efficient in its wettability between the rubber and fabric in contrast to the unfilled EPDM rubber. Increasing the filler content to 10 wt.% in case of the both regimes, poor adhesion was observed. Because this can drive to form the clay aggregation in the matrix, resulting less wettability with high interfacial tension between the rubber and the polyester fabric. This can be shown in Fig. 5. That means, the 5 % by weight of the EOC and EOC can be considered as an optimium weight.

**3.5. Flammability test**

The flammability performance of proofed samples was evaluated to be the most parameter for predicting fire hazard by measuring the rate of burning as a function of filler amounts in case of two different systems, in comparsion with unfilled proofed sample. The results are reported in Fig. 6. Unfilled proofed fabric is a fairly flammable material with high rate of burning (~ 78.4 mm/min). When adding 3 wt.% of EOC or OC filler, the rate of burning dramatically reduces to ~ 21.3 and 26.7 mm/min for both systems respectively. This decrease is related to clay barrier effect that acts as a shield layer to prevent the across of heat transfer and oxygen diffusion into the core of the material [28,29]. On further addition of filler content to the matrix, a slight raise in the rate of burning for the both systems was observed, as shown in Fig. 6. This was an expected results bescause of the excess of stearic acid or organomodifier between the clay layers that could accelerate the flammability rate at high filler content.



**Figure 5.** SEM micrographs for unfilled EPDM fabric and its filled proofed fabrics by various EOC and OC loads after peel tests.

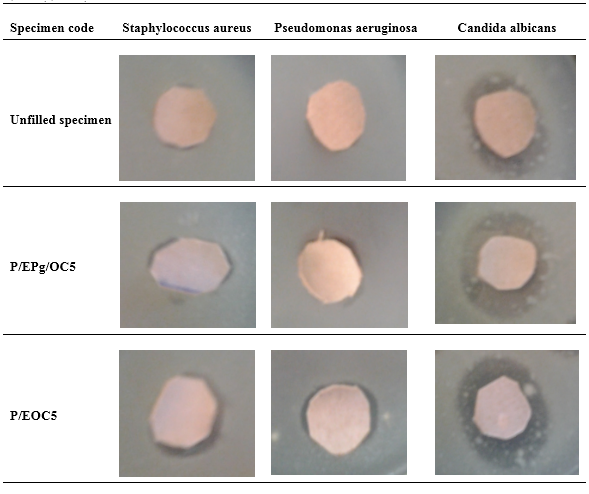
**Figure 6**. Rate of burning as function of filler contents for two proofed fabric regimes.

**3.6.** **Antimicrobial activity**

The antimicrobial test for rubber-coated fabric specimens with 5 wt.% ratio of EOC or OC containing the fixe amount 20 wt.% of LAR resin was performed to assess these fabrics for their making potentially increasing in hospital applications. The test was carried out against three different kinds of microbes: Staphylococcus aureus (G+ve bacteria), Pseudomonas aeruginosa (G-ve bacteria), and Candida albicans (fungi). The resuls are reported in Table 2 and Fig. 7 reveal that unfilled proofed sample has an inhibition effect against for G+ve bacteria (i.e 2.0 mm) and fungi (i.e 8.0 mm) used in the experiment, whereas no growth zone for G-ve bacteria in the Peri dish is observed. With introduction of 5 wt.% of OC or EOC to the sample, the inhibitory growth zone increased for all selected microbies with various diameters ranging from 3 to 17.5 mm relying on the microbial and the proofed sample type, as shown in Table 2 and Fig. 7. This improvment may be attributed to the quaternary ammonium and hydroxyl groups of organomodifier, as well as the carboxylic acid groups of stearic acid in the clay galleries that have a strong effectiveness on the microorganisms growth. Correspondingly, Zheny et al. [30] reported that long-chain unsaturated fatty acids are considered the key ingridents of antimicrobial additives Consequently, it can be concluded here that, in the presence of 20 wt. % of LAR resin (fatty acid) with EOC or OC is probably an essential component of microbial fatty acides, that served as a promosing target for antimicrobial proofed fabric applications.

**Table 2**. Antimicrobial activity of rubber-coated fabric samples against Staphylococcus aureus (ST), Pseudomonas aeruginosa (PS) and Candida albicans (CAN).

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Inhibitory zone (φmm) | | | | |  |
| Candida albicans | Pseudomonas aeruginosa | Staphylococcus aureus | Specimen | | |
| 8.0 | 0 | 2.0 | Unfilled specimen | | |
| 15.0 | 3.0 | 5.0 | P/EPg/OC5 | | |
| 17.5 | 5.7 | 8.3 | P/EOC5 | | |



**Figure 7.** Photograph of antimicrobial activity results of rubber-coated fabric specimens (unfilled specimen, P/EPg/OC5 and P/EOC5 proofed specimens).

4. Conclusions

In this research, the coated proofed fabric from EPDM/EOC to polyester fabric was prepared by spreading technique. Expanded OC by steric acid was used as wettable and reinforcing agent to obtain toxcicity-free nanocomposites with affordable cost. XRD and F-IR for OC and EOC pointed out that the d-spacing distance expanded from 1.8 nm to 4 nm, respectively. A significant improvement in the adhesion strength for P/EPg/OC and P/EOC systems was observed, especially at 5 wt.% of filler content as compared to 20 wt.% of carbon black. SEM micrographs for investigated samples after the peel test shawed that EPDM/EOC was embedded and more compatible with the polyester fabric, showing strong adhesion between them. Flammability behavior for coated proofed fabric was enhanced with adding EOC or OC as compared to unfilled sample. Poofed fabric based on EOC exhibited strong antimicrobial activities against G+ve and G-ve bacteria, as well as fungi as compared to unfilled sample or sample based on OC.

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