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REVIEW ARTICLE

Biodegradable Cellulosic Sanitary Napkins from Waste Cotton and Natural Extract Based Anti-bacterial Nanocolorants

G. Sathishkumar[1], M. Aarthi[2], R. Senthilkumar[3], P. Nithiya[4], R. Selvakumar[4] and Amitava Bhattacharyya[1]*

Abstract | With the aim to develop biodegradable quality sanitary pads at affordable prices to the schoolgirls and women, locally available cotton waste materials and anti-bacterial nanocolorants are used to prepare anti-bacterial biodegradable absorbent material suitable for sanitary napkins and diapers. Cotton fluff from loom waste was collected, cleaned and hydro-entangled to form absorbent sheet. The sheet shows water absorbency of more than 470%. Electrochemically synthesized natural extract (from neem and orange peel) based nanocolorants were incorporated in the material to improve their anti-bacterial ability. Further, microporous polylactic acid biodegradable sheet was used to pack the absorbent layer and its performance was tested. This anti-bacterial biodegradable absorbent pad would be an ideal substitute for synthetic sanitary pads considering its 100% biodegradability. This can provide a long-term viable alternative to the current practices.

1 Introduction

Affordable sanitary pads and diapers can be produced using locally available materials from anti-bacterial biodegradable cellulosic materials. Sanitary pads made from anti-bacterial biodegradable cellulosic fibre materials would also be more environmentally friendly than the synthetic non-biodegradable commercial pads. A study in 2017 showed that on an average 240 sanitary napkins are used by menstruating women each year. 10,000–12,000 disposable menstrual products are used by a woman in her lifetime. These synthetic pads take around 500–800 years to biodegrade. Sanitary napkins were disposed to open environment leading to contamination and pollution. These cannot be recycled or reused[1]. The main materials used in napkin are usually bleached rayon (pulp), cotton, superabsorbent polymer (SAP) and plastic back sheet. The bleached rayon used to absorb moisture, contains dioxin which can cause ovarian cancer. Other than dumping in a landfill, conventional hygiene products can be incinerated, but that will cause Air pollution[1]. Thus, the world is facing a very big problem of carbon footprint of feminine hygiene product as huge amount of non-biodegradable material is dumped in the landfill. This releases harmful gasses into the atmosphere[2]. In old days, cotton cloths were used for this purpose which were often reused by the rural women. This was not a hygienic practice and it was gradually replaced by the disposable synthetic counterpart. Though the disposable cotton cloths are effective to lower down gynaecological problems as compared to sanitary napkins, the absorption capacity is much higher in SAP based synthetic napkins[3]. In the modern society most of the diapers are made by synthetic materials which are non-biodegradable and have poor antimicrobial properties[4]. Looking into the problems related to these synthetic materials, researchers are focused on biodegradable super absorbent products for this purpose. Biodegradable sanitary napkins using wood fiber and cotton fabric were developed with neem (*Azadirachta indica*) and turmeric (*Curcuma longa*)[5]. However, protective films in hygiene products are mostly non-biodegradable plastic such as polyethylene and polyurethane.

[1] Functional, Innovative and Smart Textiles, PSG Institute of Advanced Studies, Coimbatore, India.
[2] Department of Chemistry, P.S.G.R. Krishnammal College for Women, Coimbatore, India.
[3] Department of Electronics and Communication Engineering, PSG College of Technology, Coimbatore, India.
[4] Nanobiotechnology, PSG Institute of Advanced Studies, Coimbatore, India.
*abh@psgias.ac.in

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This plastic material can also be replaced by bio compostable plastic prepared from starch. Many research works on bio-plastic are in progress. An alternative sustainable replacement of petroleum based polymer is poly lactic acid (PLA), derived from corn starch. It is most promising thermoplastic biodegradable polymer material. Non-toxic and bio-degradable characteristic of PLA makes it an excellent choice to be used in hygiene industry[5].

In this work, waste cotton fluffs were collected from local weaving industries and cleaned to extract the pure cotton. As this is a loom waste, it is very short length, powder like not suitable for any conventional industry recycling applications other than boiler feed. Hence, it is very cost effective. For the intended application, this material is selected because of its short size which improves its surface area leading to higher water absorbency without much after treatment. Further, its anti-bacterial ability is enhanced using natural extract (neem and orange) based nanocolorants synthesized using electrochemical method with copper electrodes[6].

Neem (*A. indica*) is in the mahogany family Meliaceae. The most important quality of Neem is less toxic to warm-blooded like human. It is having more than 300 active compounds isolated from different parts such as leaves, bark and seeds of neem tree. Neem has received a lot of attention worldwide for its prospective used as an herbal pesticide and other healthcare formulations in various countries. Neem leaves have also been used to treat skin diseases like eczema, psoriasis and rashes. Neem gel shows good anti-bacterial ability[5]. Similarly orange peel extract also shows antimicrobial activity by the agar well diffusion method against different microorganisms[7]. Copper is more abundant hence cheaper material to use. Copper nanoparticles have great applications as heat transfer systems[8] and as antimicrobial materials[9]. Copper nanoparticles have high surface to volume ratio which makes them highly reactive to interact with other particles and increase their antimicrobial efficiency. Neem and orange peel extracts have been used as capping agent during copper based nanocolorant synthesis to impart antibacterial properties in the cotton absorbent pad.

2 Materials and Methods

Waste cotton fluffs were collected from local weaving industries. The cotton was cleaned and scoured with 1 g/l sequestering agent [ethylene diamine tetra acetic acid (EDTA)], 5 g/l sodium hydroxide, 2 g/l wetting agent and 3 g/l sodium carbonate for 60 min at 90 °C with continuous stirring. All chemicals were purchased from Loba Chemie, India. After scouring, the cotton was dried and the yield was found to be $72 \pm 2\%$. The pure dried cotton fluffs were stirred with 1 l distilled water for 1 h and spread uniformly on a plastic tray. The water was squeezed out using a wire mesh to form a cotton sheet (Fig. 1). The sheet was dried in hot air oven for 3 h at 70 °C and its thickness was approximately 2 mm.

For preparation of cotton sheet with extracts, 150 ml extract (neem, orange peel extract and orange + neem extract) was added on individual samples after water squeezing from the cotton sheet and dried (Fig. 2). Freshly collected neem leaves and orange peels are crushed using grinder, filtered and lyophilized to make the extract powder. 15 g powder was dissolved in 150 ml of water to prepare the extract solution.

For preparation of nanocolorant solutions, 70 ml extract solutions were taken in the beaker with 2 g of Citric acid and 1 g of sodium chloride. The copper strip anode and cathode were immersed in the solution and voltage was applied to the solution as per Table 1. The copper ions were released from the anode which is stabilized by the extracts to form the nanoparticles expressing different colours. The anti-bacterial

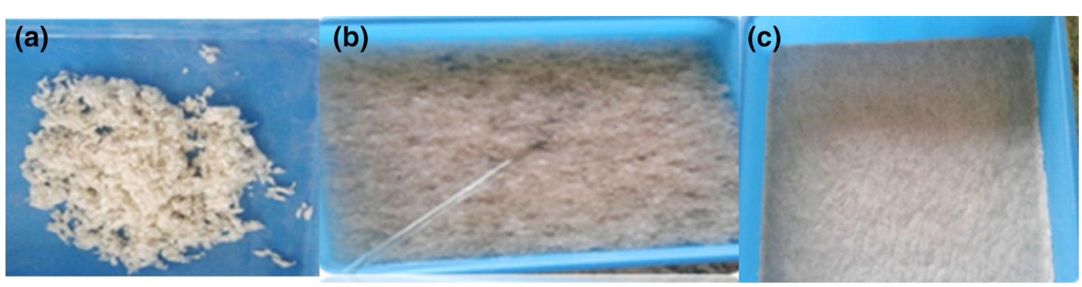

Fig. 1 Cotton ansorbent pad preparation: **a** cleaned cotton fluff, **b** cotton spread with water, **c** laid on a tray after water squeezed out

Fig. 2 **a** Neem leaves, **b** lypphillized extracted neem, **c** extracted neem solution, **d** orange peel, **e** lypphillized extracted orange peel, **f** extracted orange peel solution, **g** neem cotton sheet, **h** orange cotton sheet, **i** neem + orange cotton sheet

Table 1 Electrochemical conditions for nanocolorant formation

S. no	Sample	Current (mA)	Voltage (V)	Time taken (min)	Colour obtained
1	Neem	0.06	7.6	30	Green
2	Orange	0.06	7.8	120	Ice blue
3	Neem + orange (1:1)	0.06	7.6	60	Green

behaviour of the solution was tested and these solutions were used for the dyeing purpose.

Cotton absorbent pads were sealed with microporous PLA sheet (30 µm thickness, average pore diameter 160 µm) to develop the final structure as shown in Fig. 3.

For characterization, nanocolorants are observed under transmission electron microscope (HRTEM, Jeol JEM2100, Japan). Functional groups present in the samples were analyzed using Fourier transform infrared spectroscopy (FTIR) [Shimadzu (IRaffinity-1), Japan]

Fig. 3 **a** Microporous PLA sheet, **b** microporous PLA covered pure cotton absorbent pad, **c** microporous PLA covered cotton absorbent pad (orange cotton, neem cotton, orange + neem cotton)

at a spectral range of 4000–400 cm^{-1}. Tensile tests were carried out for cotton non-woven, microporous PLA film and PLA covered nanocolorant coated cotton absorbent pads (20 mm width) using universal testing machine (UTM, Instron 3366, USA) at 30 °C, 1 kN load cell, pneumatic grip with a 2 mm/min testing speed and 20 mm gauge length. Biodegradation study was carried out in simulated body fluid (SBF). The solution was prepared as per Gopinathan et al.[10] UV–vis spectroscopy was carried out on leached extracts under UV-1800 (Shimadzu, Japan).

The two bacteria colonies were selected for the anti-bacterial test such as *Staphylococcus aureus* (Gram positive) and *Escherichia coli* (Gram negative). The sub-culturing of the bacteria was made by preparing nutrient broth for two colonies and inoculating of bacteria in broth. The sub-cultured bacteria were taken for zone of inhibition antibacterial test. Well diffusion method was carried out by adding 50 µl of nanocolorant solution. The nanocolorant solutions of neem (a), orange (b) and neem + orange (c) were converted to different concentrations (pure (10% w/v), 7:3, 4:6 and 1:9 dilutions) for dilution effect testing of well diffusion. Further, 1 × 1 cm cotton samples dyed with nanocolorantswere also tested for zone of inhibition.

3 Results and Discussion

3.1 Nanocolorant Formation

Figure 4 shows the dispersed nanocolorants after the electrolytic deposition. Tindal effect was observed in the solutions (Fig. 4b, c) when a laser was passed through the dispersion indicating the formation of nanoparticles.

Further, the Cu-nanoparticles were analysed by transition electron microscopy (Fig. 5). The synthesized nanoparticles dimensions were found to be within 5 nm. For orange + neem solution, nanoparticles yield was higher and it is uniformly distributed in the dispersion.

3.2 FTIR

Fourier transform infrared spectroscopy (FTIR) identifies chemical bonds in a molecule by producing an infrared absorption spectrum (Fig. 6a).

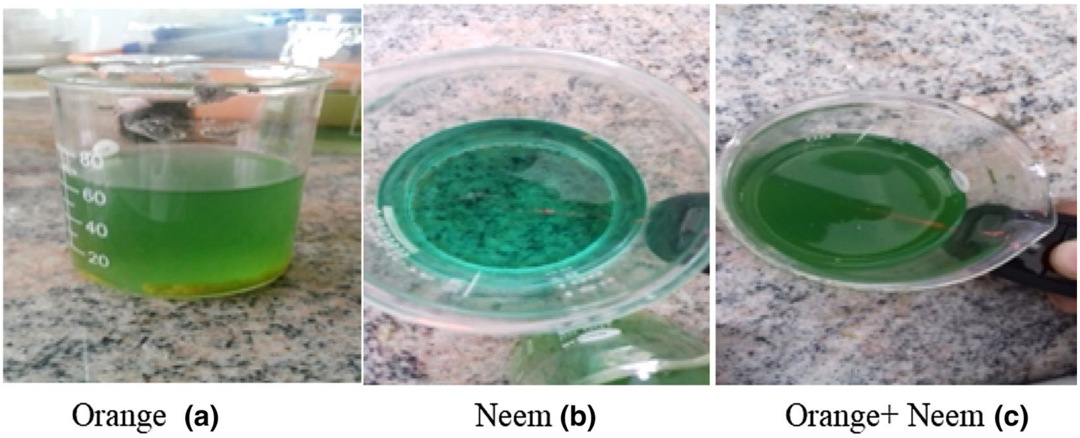

Fig. 4 **a** Orange to lemon green, **b** dark to light green and **c** yellowish green to light green

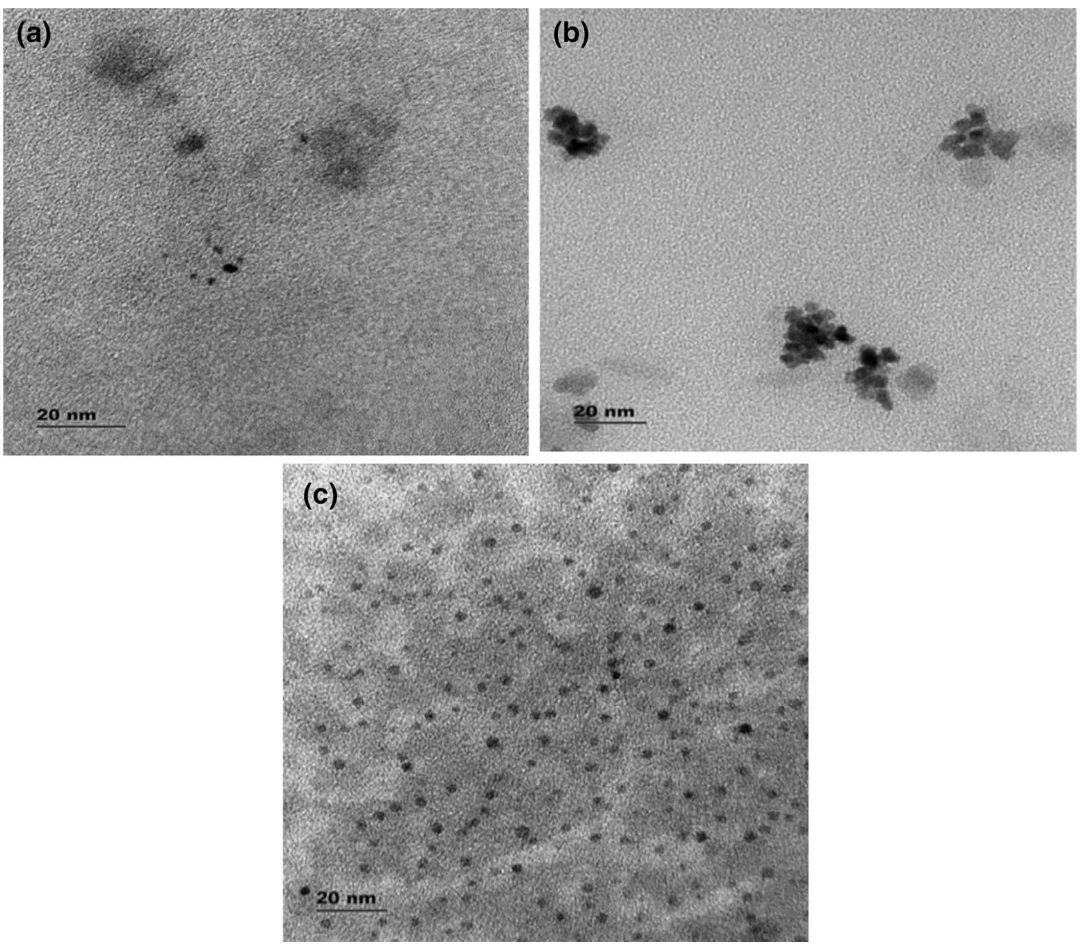

Fig. 5 TEM images of the nanoparticles: **a** orange Cu-nanocolorant solution, **b** neem Cu-nanocolorant solution, **c** neem + orange Cu-nanocolorant solution

The spectra produce a profile of the sample, a distinctive molecular fingerprint that can be used to screen and scan samples.

The band at 1160 cm^{-1} was assigned to bond of C–H wagging vibrations. It has alkyl halides functional group. 1055 cm^{-1} was assigned to

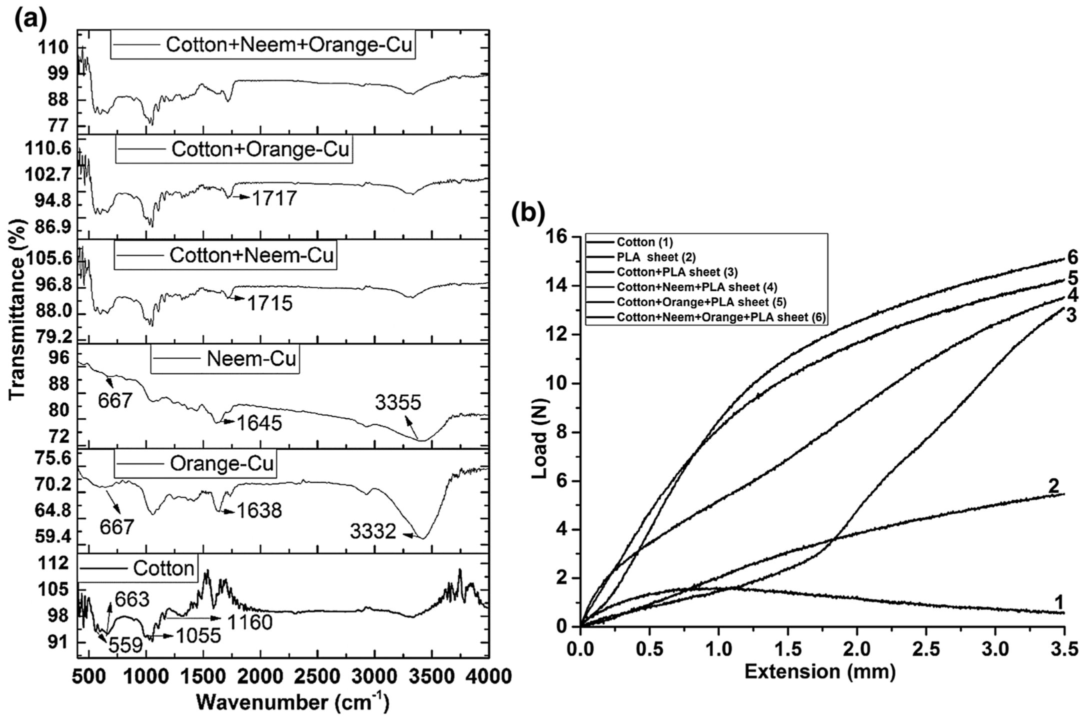

Fig. 6 **a** FTIR spectra of cotton, nanocolorants and nanocolorant coated cotton, **b** tensile test load-elongation curves of cotton, PLA sheet and PLA sheet covered absorbent pads

bond of C–N stretch. It was related to the maximum functional group of the aliphatic amines. For the orange peel extract mediated copper nanoparticles, the peaks at 3332 cm^{-1} was assigned to bond of N–H stretch (3400–3250 cm^{-1}) and O–H stretch (3500–3200 cm^{-1})[11]. The peaks at 1638 cm^{-1} was denoted as bond of N–H bend (1650–1580 cm^{-1}) related to the functional group of primary amines. The peaks at 667 cm^{-1} was assigned to the functional group of alkynes (700–610 cm^{-1}). This peak was strong and base peak in Cu-nanocolorant of orange solution. The peaks at 1717 cm^{-1} was assigned to bond of C=O stretch[12]. This is a strong peak observed in orange Cu-nanocolorant dyed cotton.

For the neem-based copper nanoparticles, the peaks at 3355 cm^{-1} was assigned to bond of N–H stretch (3400–3250 cm^{-1}). It was related to the functional groups of the primary and secondary amines, amides and O–H stretch. These consist of maximum functional group of alcohols, phenols. The peaks at 1645 cm^{-1} was assigned to bond of N–H bend (1650–1580 cm^{-1}) i.e. functional group of amines. The peaks at 667 cm^{-1} was assigned to the functional group of alkynes (700–610 cm^{-1}). This peak was strong in Cu-nanocolorant of neem solution. The peaks at 1717 cm^{-1} was assigned to bond of C=O stretch (1730–1715 cm^{-1}). It was related to functional group of α, β unsaturated esters. This is obtained for both orange and neem Cu-nanocolorant of dyed cotton.

3.3 Tensile Tests

Figure 6b shows the tensile test curves of cotton absorbent pad, microporous PLA sheet and the four different absorbent pads with nanocolorant sandwiched between two layers of microporous films. The multi layered structure provides structural stability to the absorbent pad, so the tensile properties have been improved for the sandwich structures (curve 3–6). Use of nanocolorants further improved the tensile properties up to 14 N with 3.5 mm extension (~ 17.5%) which indicated that the structural integrity was enhanced for those pads. The load extension values confirm that the multi-layered pad is structurally stable to withstand reasonable amount of load during the conventional use.

3.4 Biodegradation Study

Degradations of different materials after 30 days are shown in Table 2. Raw neem extract based material degrades slowly while both pure cotton and orange extract based materials showed high

Table 3 Water absorbency properties of components used for making biodegradable sanitary napkin

Sample	Absorbency (%)
Pure cotton	470 ± 5%
Neem cotton	220 ± 5%
Orange cotton	271 ± 5%

degradation within 1 month. Pure cotton results more than 50% weight loss within 30 days.

Figure 7 shows UV–vis absorption spectra of degradation media after extract released. Both neem and orange extract leached in the solution during degradation as the intensity increases with time. The peak intensity of orange based sample increased more as the degradation is faster in orange cotton sample.

3.5 Absorption Capacity of Sanitary Napkin

Absorbency of napkin depends more on the core material than the surface layer. There is significant difference between the samples treated with raw extracts (Table 3). The SAP used in commercial sanitary napkins can absorb up to 300 times on its own weight. The use of SAP in the napkin inner absorbent layer is restricted and it is usually mixed with cotton, viscous fluff or encapsulated with polypropylene because of its poor feel after absorption[13]. Still the water absorption capacity of the absorbent layer of commercial non-biodegradable products is much higher than the developed one. However, the significance of this work lies on reusing the cotton waste as well as preparing a 100% biodegradable sanitary napkin.

It has been observed that the raw extract reduces the water absorbency from 480 to 270%. Further, with addition of PLA sheet the total product absorbency goes down, as PLA is not participating in water absorption (pure cotton: 189 ± 5%, neem cotton: 113 ± 5%, orange cotton: 157 ± 5%).

Hence, nanoparticle based anti-bacterial agents has been studied to improve the anti-bacterial ability while retaining the water absorbency. The amount or dosage of extract used in nanoparticles form would be much less compared to the raw extract treated samples.

Table 4 shows the water absorption of different cotton pads with nanocolorants. Orange nanocolorant shows good absorbency. All samples show better absorbency than the raw extract.

Table 2 Biodegradation in presence of simulated body fluid

Sample	Initial weight (g)	Final weight after 30 days (g)	Degradation (%)
Pure cotton	0.2816	0.184	53
Neem cotton	0.2897	0.2111	27
Orange cotton	0.3169	0.2173	45

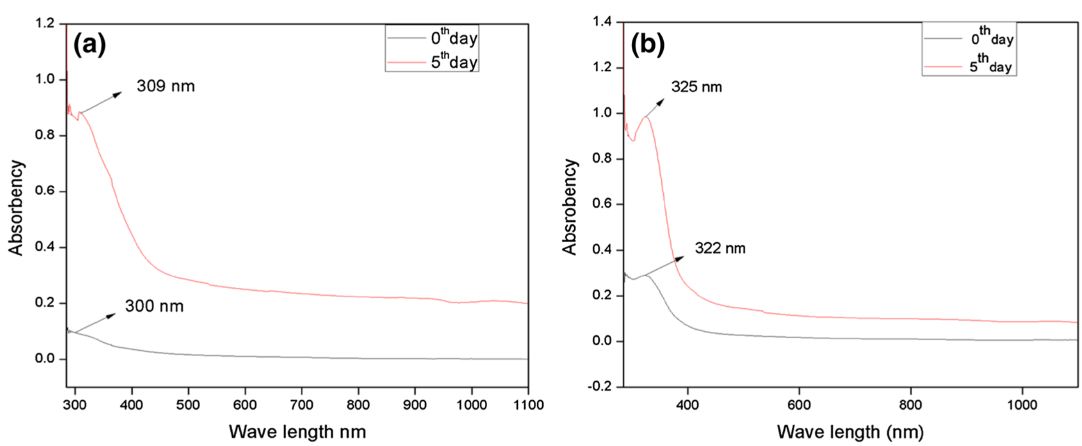

Fig. 7 Extract releases in degradation media (0th day and 5th day) **a** neem, **b** orange. Showing the absorption spectra of the solution media on 0th day and 5th day

Table 4 Water absorbency properties of cotton pad with Cu-nanocolorant components without PLA sheet used for making biodegradable sanitary napkin

S. no.	Sample (3 x 5 cm)	Absorbency (%)
1	Neem cotton	263 ± 5%
2	Orange cotton	308 ± 5%
3	Neem + orange cotton	270 ± 5%

3.6 Assessment of Antimicrobial Activity

The anti-bacterial tests carried out with the raw extracts have not shown successful results. The copper nanoparticles was biologically synthesized using plant leaf extracts of Meliaceae (*A. indica*), and Rutaceae (*Citrus sinensis*) as reducing and stabilizing agents. On treatment of copper electrode with the leaf extract and orange peel extract stable copper nanoparticles were formed. The antimicrobial activity of *C. sinensis* and *A. indica* is evaluated using qualitative well diffusion method as shown in the Fig. 8. The results clearly show that the treated sample with Cu nanocolorant of *C. sinensis* and *A. indica* show good antimicrobial activity with Gram-positive organism, the inhibition region was found to be 18 mm and for Gram-negative organism it shows no inhibition region. The control specimen shows no zone of inhibition for both Gram-positive organism and Gram-negative organism. A zone of inhibition

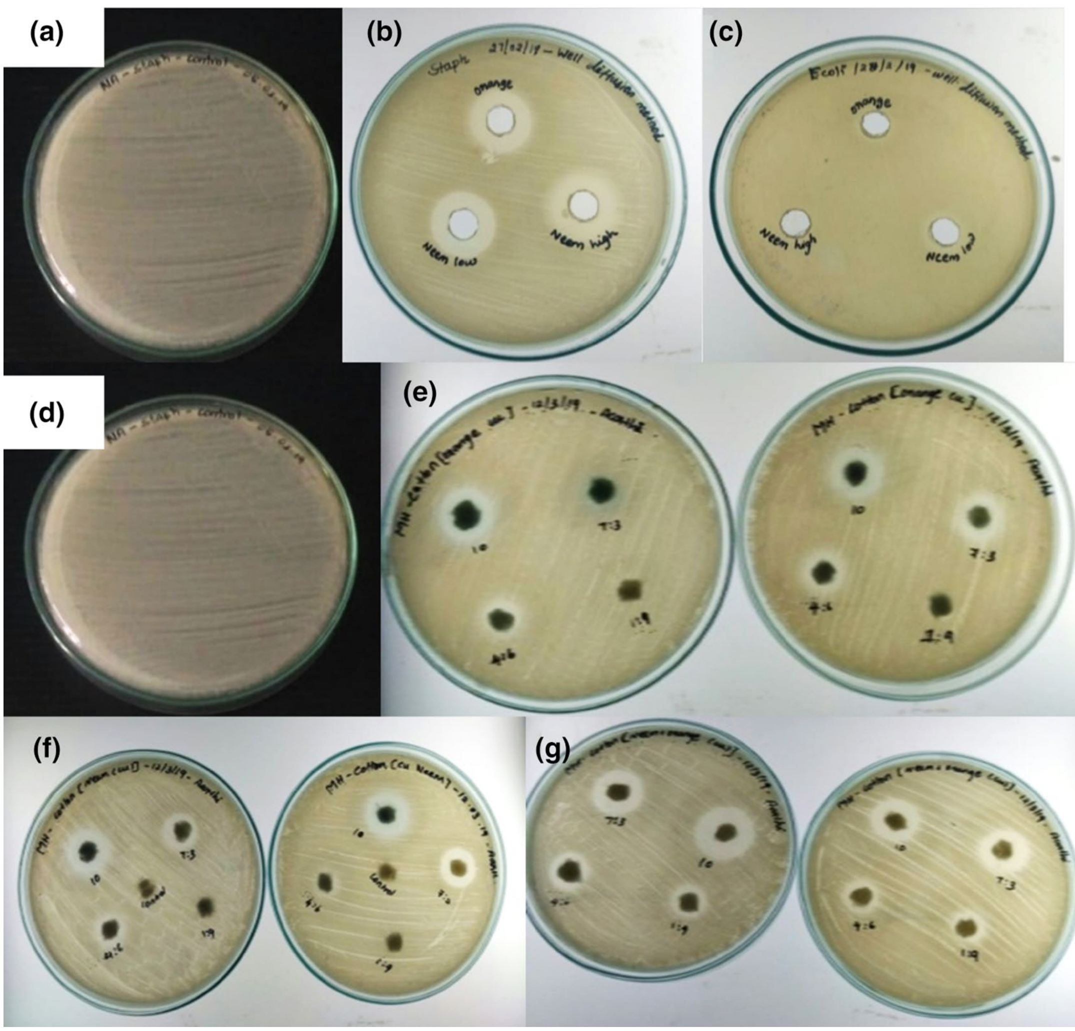

Fig. 8 Anti-bacterial test in well diffusion method: **a** control, **b** 50 µl Cu-nano colorant solution (orange and neem) of *Staphylococcus aureus*) (gram positive), **c** 50 µl Cu-nano colorant solution (orange and neem) of *E. coli* (gram negative). Test with different concentrations: **d** control, **e** 1 Cu-nanocolorant neem cotton (6 mm), **f** 1 Cu-nanocolorant orange cotton (6 mm), **g** 1 Cu-nanocolorant neem + orange cotton (6 mm) of *Staphylococcus aureus* (gram positive)

Table 5 Zone of inhibition (mm) at different dilutions

Sample	Dilutions (mm)			
	Pure (10% w/v)	7:3	4:6	1:9
Neem	18	14	13	–
Orange	17	15	14	–
Neem + orange	20	17	14	12

occurs as a result of the diffusion of an antimicrobial agent from the specimen. For nanocolorants, clear area of no growth of microorganism was observed for the bacteria cultured on to the surface of an agar growth medium, in proximity to the borders of a specimen placed in direct contact with this agar surface.

In Fig. 8b, added 50 µl of nanocolorant solution in each hole showed same zone of inhibition level (18 mm). From Fig. 8d–g, it is concluded that when the concentration was increased, the zone of inhibition level also increased. Table 5 shows the zone of inhibition at different dilutions of nanocolorants. Neem orange combination at very low dilution is also shows good antibacterial ability. Hence, this nanocolorant can be employed in such biodegradable cotton pad to impart good antibacterial ability as well as reasonably high biodegradation after exposure to external world.

4 Conclusion

As the same cotton lint was used as raw material to make non-woven sheets using same technique, structurally all are similar. However, due to the use of neem and orange peel raw extract, its water absorbency reduces from 480 to 270%. The extracts did not contribute on water absorbency, rather they impart antibacterial properties. Most of the cotton absorbency can be retained using nanocolorants, because the amount added to the cotton lint is much less though its antibacterial properties are good. Use of nanocolorants significantly improves the water retention capacity of the extract treated anti-bacterial absorbent cotton pads. Further, its structural integrity was also improved as evident from the tensile test results. With a PLA sheet enclosure, 100% biodegradable, anti-bacterial product can be prepared using this method. Both the antimicrobial finishes Meliaceae (*A. indica*), and Rutaceae (*C. sinensis*) show good antimicrobial activity against gram positive bacteria. The concentration can be increased for increased zone of inhibition. Combined use of neem and orange peel extract based nanocolorant was resulted in good antibacterial, tensile and absorbency property with moderate degradation. Though the water absorption capacity of commercial non-biodegradable sanitary products is much higher than the developed one, this work stands significant considering the reuse of cotton waste to prepare a 100% biodegradable sanitary napkin. As the product is biodegradable, it will prevent non-biodegradable waste generation. Sanitary napkins should not only provide comfort and safety but must enhance woman's health and hygiene. This technology has scope in the future because of the personal hygiene as well as environmental protection.

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Mr. G. Sathish Kumar is presently working as a Junior Research engineer in FIST Lab at PSG Institute of Advanced Studies, Coimbatore, Tamil Nadu, India. His major research interests are polymer nanocomposites and electrospun functional nanofibers.

Ms. M. Aarthi is presently pursuing B.Ed. course in Ponmari college of Education in Narthamalai, Tamil Nadu, India. She has completed Master of general chemistry at PSGR Krishnammal college for women in Coimbatore. Her major research interests are nanotechnology and nanomaterial synthesis.

Mr. R. SenthilKumar is presently a Senior Research Fellow in the project titled "Development of smart socks for wireless monitoring of human gait" and pursing Ph.D. in polymer nanocomposites from PSG College of Technology, Coimbatore, Tamilnadu, India. His major research interests are wearable sensor, biosensors, biomedical devices 3D printing, and nanotechnology.

Ms. P. Nithiya completed her M.Sc. and M.Phil. in nanoscience and technology in Bharathiar University and currently she is pursuing her Ph.D. in nanobiotechnology at PSG Institute of Advanced Studies, Coimbatore, India. Her major research interests are nanobiotechnology, nanomaterials for environmental applications and emerging contaminants in water.

Dr. R. Selvakumar is presently working as Associate Professor at PSG Institute of Advanced Studies, Coimbatore, Tamil Nadu, India. His research interests include nanobiotechnology, tissue engineering, biosorption and microbial biotechnology with specialization on microbial-metal interactions, and nanoparticle-based environmental technology.

Dr. Amitava Bhattacharyya is an Associate Professor at PSG Institute of Advanced Studies, Coimbatore, Tamil Nadu, India. His major research interests include high-performance polymer nanocomposites, biomedical devices, electrospun functional nanofibers, and designing of polymer scaffolds for tissue engineering.