RJC

RASĀYAN *J. Chem.*
Vol. 14 | No. 1 |382-388| January - March | 2021
ISSN: 0974-1496 | e-ISSN: 0976-0083 | CODEN: RJCABP
http://www.rasayanjournal.com
http://www.rasayanjournal.co.in

DEVELOPMENT OF ANTIBACTERIAL COTTON FIBERS BY APPLICATION NANOPARTICLE PRODUCED BY MANGOSTEEN RIND

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ABSTRACT

A silver nanoparticle is one material that has antibacterial activity. The objectives of this study are to determine the contact angle and antibacterial activity of cotton fiber by adding Ag nanoparticles and hexadecyltrimethoxysilane compound. The Ag nanoparticle was produced by the active component of mangosteen rind (*Garcinia mangostana* L.) as a natural reducing agent. The addition of hexadecyltrimethoxysilane (HDTMS) compounds is also done to make cotton fibers are hydrophobic. The Ag nanoparticle was performed by using Ultra Violet - Visible spectrophotometer, the antibacterial activities of cotton in inhibiting *S. aureus* and *E. coli* were analyzed by measuring an inhibition zone, and hydrophobicity properties of cotton before and after modification were measured using the method of the sessile drop. The colloid of Ag nanoparticles was produced at 442 nanometers and have an average diameter size of as much as 116 nanometers. The cotton after modification with HDTMS showed the biggest contact angle as much as 129.55°. The cotton after modification with HDTMS and Ag nanoparticle has the highest inhibition activity against *S. aureus*. These cotton fibers can be used as antibacterial products with a strength 4 to 5 times greater than in silver nanoparticles.
Keywords: Antibacterial Activity, Cotton, HDTMS, Mangosteen Rind, and Nanoparticle.
RASĀYAN *J. Chem.*, Vol. 14, No.1, 2021

INTRODUCTION

Cotton fiber is one of the natural fibers that are widely used in the textile industry. It's convenient when used to make cotton fiber is very popular. However, behind its good nature, cotton fiber is not sterile and a good medium for the growth of microorganisms because of its properties that can retain moisture[1]. Therefore, modifications should be made to improve the quality of the cotton fibers. The development of nanotechnology in the field of textiles is currently very rapid. This is demonstrated by the emergence of higher-performance textile products such as extreme heat-retarding clothing, textiles with self-cleaning textile properties, antimicrobial textiles that can be used in the medical and military world. Applications of nanotechnology in textile materials can be done by depositing nanometer-scale particles into textile fibers.[2,3] Silver colloids have long been known to have antimicrobial properties.[3-5] The process of making silver nanoparticles can be done in several ways. One way is by the method of reduction. Currently, there have been many studies that use chemical reducers as reducing agents to produce silver nanoparticles. However, chemical compounds can also adversely affect the environment, such as environmental pollution. Therefore, in this study, the production of Ag nanoparticles is done via the reduction method with natural ingredients as a reducing agent or so-called bioreduction. Excess use of natural ingredients as a reducing agent is environmentally friendly, easy to obtain, and also simple.

Based on research that has been done, many plants can be used for the preparation of silver nanoparticles, including Ketapang leaves[6], betel leaves[7], cinnamon, red fruit, *Ipomoea batatas* L.[8], mangosteen rind, and so on. The plant used in this study is the rind of the mangosteen fruit (*Garcinia mangostana* L.). Mangosteen rind is extracted and used as a natural reagent in the preparation of Ag nanoparticles.

The hydrophobic properties of cotton fibers can be obtained by adding silane-based molecules such as Methacryltrimethoxysilane, Octyltriethoxysilane, Methyltriethoxysilane, Hexadecyltrimethoxysilane, and Trimethylethoxysilane.[3, 9] The resulting fiber becomes a self-cleaning textile because it has a property known as the lotus effect that is like the surface of Lotus leaf (*nelumbo nucifera*) which is hydrophobic.The nature of the leaf hydrophobes is caused by the complex surface texture of the micro-to-nano scale. When

http://dx.doi.org/10.31788/ RJC.2021.1416034

the leaves splash water then the water will form the grains on the leaf surface and immediately fall when the leaf is tilted. The nanostructured three-dimensional surface particles and gel-producing additives produce hydrophobic fiber products without reducing the comfort of the fiber when worn. Cotton fibers modified with the addition of silane compounds will have very low surface energy.[10]
Methods to improve the stability of properties and hydrophobic in cotton cloth developed in this study. The silver nanoparticle-derived fibers will produce a coarse surface texture, then modified with the Hexadecyltrimethoxysilane (HDTMS) chemically bound to the particles of the material through surface condensation reactions. The addition of HDTMS compounds to nanoparticle-loaded cotton fiber is expected to develop the quality of cotton to be hydrophobic and antibacterial.

EXPERIMENTAL

Materials

The materials used in this study, included cotton cloth, $AgNO_3$, nutrient agar, nutrient broth, polyvinyl alcohol (PVA), C_2H_5OH, acetone, and hexadecyltrimethoxysilane. Cotton cloths as a commercial product were bought from the fabric shop in Yogyakarta, Indonesia.[3] Nutrient Agar (NA) and Nutrient Broth (NB) were bought from Oxoid and used without any further purification.[5-7] *S. aureus* ATCC 25923 and *E. coli* 32518 were available in a collection of Medicine Faculty, UGM, Indonesia.[3]
The experiment was conducted with the following procedures: extracting of mangosteen rind, preparation of Ag nanoparticle, depositing of Ag nanoparticle toward cotton fibers, modification of cotton fibers with the addition of HDTMS, and characterization of cotton fibers.

Application of *Garcinia mangostana* L. to Produce Silver Nanoparticle

As much as 400 mL of distilled water was used to wash 40 grams of *Garcinia mangostana* L. rind, then 200 mL of distilled water was mixed with *Garcinia mangostana* L on boiling for about 20 minutes. It was then filtered out by using Whatman No. 42 at 270C. As much as 80 mL of $AgNO_3$ solution 0.001 mol/L and 2 mL of the extract were mixed. The mixture was allowed to react for about 120 minutes and next as much as 24 mL of polyvinyl alcohol solution 1% (v/v) was poured into that solution (extract + $AgNO_3$) and stirred for about 120 minutes. The produced solution on the previous stage was then allowed to stand for 96 hours to produce a colloidal Ag nanoparticle. The colloidal Ag nanoparticles were characterized by a spectrophotometer of UV-Vis and Particle Size Analyzer (PSA).

Impregnation of Ag Nanoparticle on Cotton

A cotton cloth with a size of 5 cm x 5 cm was washed with acetone for 30 minutes, then rinsed using nonion distilled water nonion through immersion for about 30 minutes and then dried up by hairdryer. Furthermore, cotton fiber was impregnated by immersing in the colloid of Ag nanoparticles and shaking with a shaker at 153 rpm for 24 hours and finally evaporated at room temperature.

Application of HDTMS on Cotton

Modification of the cotton and the cotton after deposited with silver nanoparticles was conducted by immersing those in the HDTMS solution.[3] The produced mixture was allowed to react for about 6 hours at room temperature. Cotton and cotton after modification with Ag nanoparticles were immersed in HDTMS solution and were twisted using a shaker at 155 rpm for 1 hour followed by drying at 80 °C for 10 minutes. Then, the curing process at 130ºC for 1 hour. Cotton fibers were characterized by measuring the inhibition zone against growth bacteria and hydrophobicity test. The fibers which were produced in this study were labeled as the followings, cotton fiber (C0), cotton fiber - silver nanoparticles (C1), cotton fiber - HDTMS (C2), cotton fiber – silver nanoparticles – HDTMS (C3), and cotton fiber - HDTMS - silver nanoparticles (C4).

Test of Antibacterial Activity

Inhibition activity against bacteria was analyzed by preparing nutrient agar and nutrient broth as media to bacteria growth as shown in previous study.[3,5-7] Every cotton sample before and after modification was prepared by cutting it using a paper hole diameter of 6 mm, then put in the Petri dish and allowed to incubate for 1440 minutes (24 hours), then was measured an inhibition zone to 4320 minutes (72 hours). The inhibition zone diameter of cotton fiber against *E. coli* and *S. aureus* were analyzed by using a statistic test

using the program of SPSS version 15. The effects of sample type, incubation time, and interaction between samples and incubation time on inhibition activity were analyzed by the ANOVA test.

Test of Water Contact Angle (WCA)

The WCA of the cotton fibers samples without and with modification was analyzed by observing the formed angle (θ/tetha) between the water and the cotton sample surface using sessile drop method.[3,5,6] Based on the images, the water contact angle was determined with the Corel Draw X4 version.

RESULTS AND DISCUSSION

Properties of Silver Nanoparticles

The extracts of *Garcinia mangostana* L.rinds and silver colloidal silver nanoparticles in this synthesis are shown in Fig.-1.The leaves extract after the addition of silver nitrate solution changes from colorless to dark brown. The changing of solution color due to the -OH group in the tannin as the main component of mangosteen rind extract has oxidized to C the functional groups, i.e. C = O groups. It indicates that the reduction process of silver nitrate solution has occurred and formed colloidal silver nanoparticles.

0 hour (a) | 1 hours (b) | 24 hours (c) | 48 hours (d) | 96 hours (e)

Fig.-1: Colloidal of Silver Nanoparticle

The analysis using UV-Vis spectrophotometer was conducted at the wavelength range 190-500 nm for $AgNO_3$ solution and the analysis of colloidal silver nanoparticles (Fig.-2).

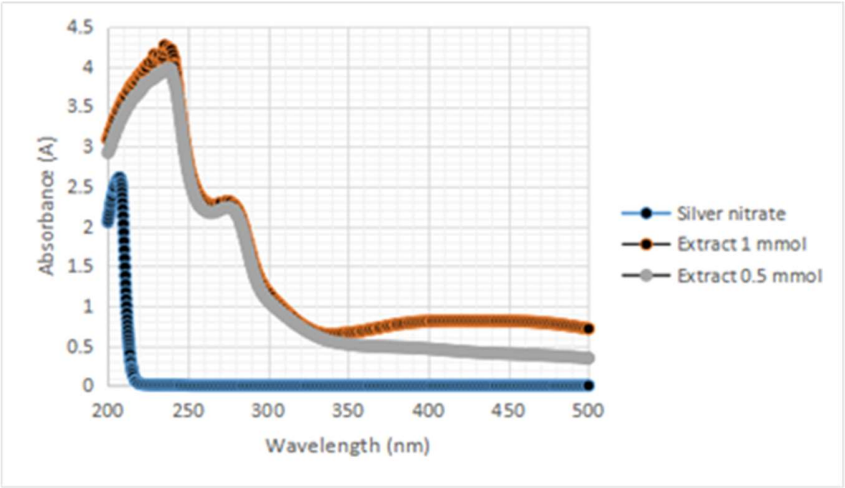


Fig.-2: The UV-Vis Spectral Patterns of $AgNO_3$ Solution and Silver Nanoparticle by Using Extract of Mangosteen Rind

The $AgNO_3$ solution shows the absorption band peak at a wavelength of 208.00 nm. While the silver nanoparticles show three-band peaks. The first peak appears at 240.00 nm, while the second band peak that appears at 282.00 nm and the third band peak at 442.00 nm show that there is still Ag^+ which could not be reduced by rind extract to Ag^0. The third band peak that appears at a wavelength of 442 nm indicates that Ag^+ has been successfully reduced to Ag^0. Tannins are compounds in the rind extract of *Garcinia mangostana* L that are thought to play an important role as a reducing agent (Fig.-4).

The silver nanoparticle has successfully been synthesized by using mangosteen rind extract. It is proved by the appearance of a band peak at 442 nm. Many kinds of literature described that nanoparticles have band peaks around 400 nm[3, 5, 6, 7]. The distribution of particle size of Ag nanoparticles is showed in Fig.-3. The average diameter of the silver nanoparticle by using mangosteen extract is 116 nm.

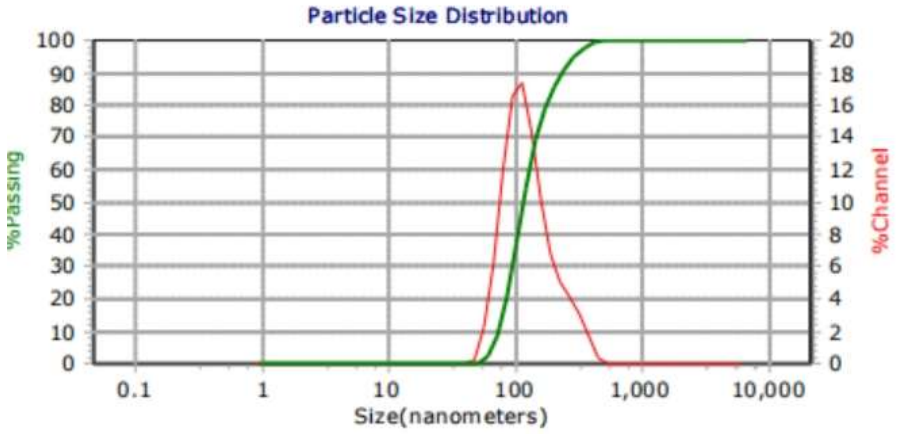


Fig.-3: The Particle Size Distribution of Colloidal Silver Nanoparticles

The mechanism for reducing silver ions to silver nanoparticles can be explained as follows (Fig.-4). The -O-H group in the structure of tannins (mangosteen extract) reacts with silver ions from the silver nitrate solution to form the -O-Ag group. Furthermore, the release of methyl groups and silver ions to form carbonyl groups (C = O) as an oxidation product of tannins and silver nanoparticles as well as solutions of nitric acid and methylnitrate.

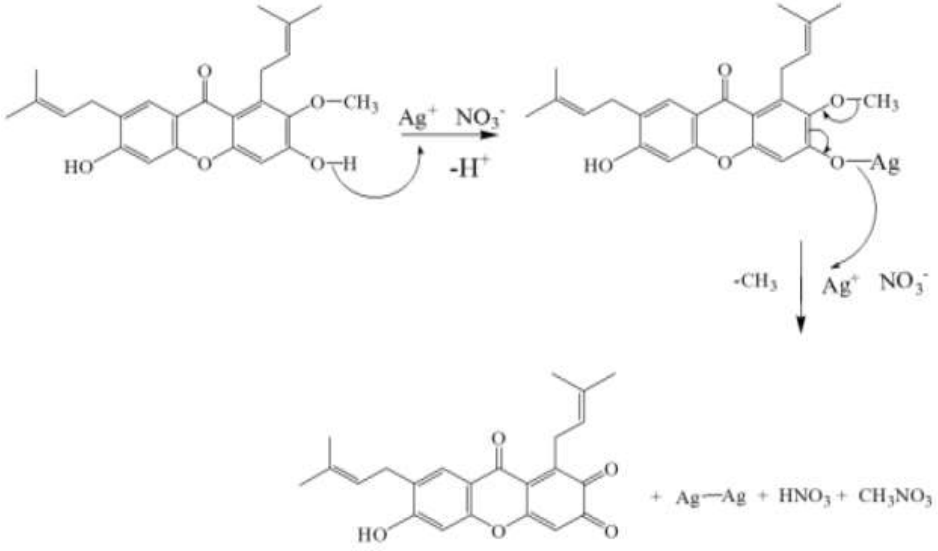
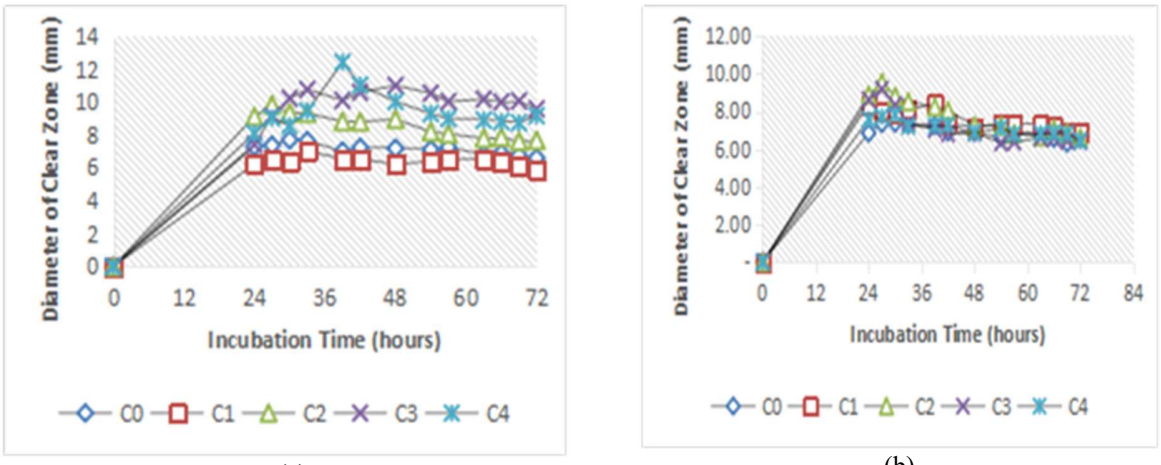


Fig.-4: Reduction of $AgNO_3$ to Silver Nanoparticle by Tannin Compound[11]

Inhibition Activity of Cotton against the Growth of *Staphylococcus aureus* ATCC-25923 and *Escherichia coli* ATCC 35218

Figure-5 shows the inhibition zone formed of cotton fibers against *S. aureus* ATCC-25923 and *E. coli* ATCC 35218. The sample of C4 shows the biggest inhibition activity (the diameter of inhibition zone = 12.43 mm) compared to other samples at 39 hours of incubation. This suggests that the addition of silane solution and also Ag nanoparticles increase the inhibition activity of cotton fibers against *Staphylococcus aureus*. Sample C2 shows the biggest inhibition activity (the diameter of clear zone = 9.53 mm) compared to other fiber samples at 27 hours of incubation. This suggests that the modification with HDTMS increases

the inhibition zone of cotton fibers against the life of *Escherichia coli*. Comparing with previous research, sodium citrate produced silver nanoparticles demonstrated a smaller width or diameter of the clear zone (2.3 mm) against *E. coli*[11]. Thus both cotton samples (C4 and C2) have much higher antibacterial activity than silver nanoparticles. Whereas silver nanoparticles are a good antibacterial ingredient.[12] Both samples can be used as antibacterial agents with a strength 4 to 5 times greater than silver nanoparticles as a positive control.



(a) (b)

Fig.-5: The Inhibition Activity of Cotton Fibers against *S. aureus* (a) and *E. coli* (b)

HDTMS compounds have hydroxyl groups so that the hydroxyl group can interact with the phosphate group in the cell wall of bacteria and cause the growth of bacteria to be inhibited. The effect of HDTMS compounds in inhibiting bacterial growth may also reduce the surface tension as well as detergents. Detergents containing hydrophilic and hydrophobic groups are antibacterial agents that can kill bacterial cells.[8,13]The silver nanoparticles inhibit and destroy a microorganism through a mechanism, where silver nanoparticles enter into bacterial cells, this causes the formation of areas with a low molecular weight in the middle of a bacterial blob that serves to protect DNA. Furthermore, the silver nanoparticles conduct diffusion and attack the bacteria's respiratory chain, until eventually, the cell becomes dead.[14]The ANOVA result is presented in Table-1.

Table-1: Two Way ANOVA: Variety of Cotton and Treatment Time against Growth of *S.aureus* and *E.coli*

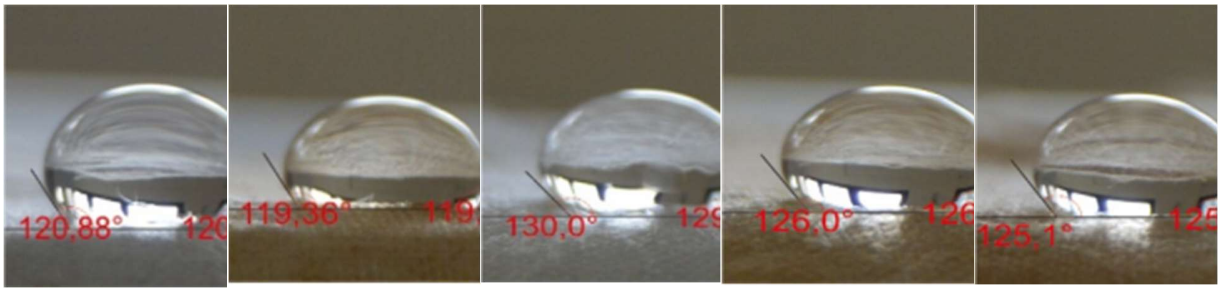
Source	Squares Sum	Free Degree	Square of Mean	F Value	Significance
S. aureus					
Incubation Time	0.347	12	0.029	6.519	0.000
Cotton Fiber	3.541	4	0.885	199.681	0.000
Incubation* Cotton	0.693	48	0.014	3.256	0.000
E. coli					
Incubation Time	0.643	12	0.054	9.137	0.000
Cotton Fiber	0.187	4	0.047	7.969	0.000
Incubation* Cotton	0.230	48	0.005	0.818	0.784

Each of the treatment times and the variety of cotton affects the inhibition activity of the cotton sample against *S.aureus* and *E.coli*. Nevertheless, the time of incubation and the type of cotton cloth did not simultaneously affect the inhibition activity of the cloth against the growth of *E.coli.* The result of LSD test showed that all cotton fibers have a significant difference in antibacterial activity against the growth of *S.aureus*, while between C0 – C3, C0 – C4, C1 – C2, and C3 - C4 do not show a significant difference in antibacterial activity against the growth of *E. coli*. Nevertheless, all types of cotton fibers in this study show antibacterial activity, so the possibility of modified cotton with HDTMS and silver nanoparticles can be used as an antibacterial agent which will protect and prevent infection and accelerate the process of wound

healing. The mechanisms of antibacterial nanoparticles include the induction of oxidative stress, the release of metal ions, and also the mechanisms of nonoxidation.[10,15]

The Water Contact Angle of Cotton Fibers

Sample C2 shows the highest contact angle (129.55^0), this can be due to the number of intramolecular hydrogen bonds in the cotton – HDTMS at most compared to the other samples. Hydrogen bonding affects the wettability of materials.[16] The HDTMS solution clearly can enhance the water contact angle of cotton. The HDTMS compound is an amphiphilic molecule with a hydrophilic head section, ($Si(OCH_3)_3$), and a tail portion which is a hydrophobic long alkyl group ($C_{16}H_{33}$). The HDTMS compound that was coated on cotton fiber interact and form a hydrogen bond intramolecular that results in free surface energy down so that the surface of the fiber will be hydrophobic. Initially, the HDTMS compound on a material surface will be hydrolyzed and produce the -OH group. The -OH group of HDTMS will form a bond with a typical group of cotton fiber surfaces i.e -OH forming Si-O-bonds. As a result of the bonding, the tail of the HDTMS is a long hydrophobic alkyl group that extends outward and becomes a barrier for water molecules soaking the fiber surface so that the cotton fiber will be hydrophobic.



θ = 120.45°	θ = 119.64°	θ = 129.55°	θ = 126.15°	θ = 125.1°
(a)	(b)	(c)	(d)	(e)

Fig.-6: The Water Contact Angle of Cotton (a), Cotton - Ag (b), Cotton - HDTMS (c), Cotton - Ag - HDTMS (d), and Cotton -HDTMS - Ag (e)

The addition of silver nanoparticles to a cotton fiber causes a decrease in contact angle (Fig.-6). The decrease of the contact angle value of cotton - Ag due to silver nanoparticles on the sample surface could be caused by the NPAg impregnated on cotton fiber have a nanometer size and the smaller the size of a material the smaller the contact angle would be. However, the contact angles of cotton fibers after modification with HDTMS is highest. The HDTMS compound causes the material dislike to water. This is caused by the tail section of the HDTMS compound which is hydrophobic, consists of a long alkyl group ($-C_{16}H_{33}$). Other than that, silane compounds act like a detergent, a long alkyl group in a hydrophobic section that would cover cotton fibers linking with Ag nanoparticles. The structure of HDTMS compound is similar to the surfactant structure. In this study, the HDTMS can act as a stabilizer of nanoparticles and cause self-cleaning of cotton fiber. The HDTMS can act as a surfactant and a stabilizer of nanoparticles.[17] The hydrophilic section in the HDTMS compound decreases the ΔG value on the surface of the cotton, so the cotton sample can work optimally.[18] The presence of surfactants such as PEG 6000 will affect the size and character of the produced nanoparticles.[19] Based on the results indicated that Ag nanoparticles deposited in cotton can cause cotton to have antibacterial properties. Thus, it is proven that nanoparticles have a function as an antibacterial material. This is in line with previous research that nanoparticles are antibacterial materials.[20,21] The hydrophobicity of the cotton fiber after modification with silver nanoparticles and HDTMS (C3) and also the hydrophobicity of the cotton fiber after modification with HDTMS and silver nanoparticle (C4) are lower than the hydrophobicity of cotton fiber after modification with just HDTMS. This shows that silver nanoparticles can decrease the hydrophobicity characteristic of cotton. The colloidal Ag nanoparticle has a hydrophilic characteristic.[22]

CONCLUSION

Colloidal silver nanoparticles were successfully prepared using the extract of mangosteen rind with a diameter size of 116 nm. Based on the analysis using a UV-VIS spectrophotometer, the colloids of silver

nanoparticles were produced successfully at 442 nanometers. The cotton after modification with HDTMS showed the highest contact angle, as much as 129.55°. The cotton – HDTMS – silver nanoparticle (C4) showed the biggest inhibition activity against the growth of *S. aureus* and the cotton – HDTMS (C2) showed the biggest inhibition activity toward the growth of *E. coli*. Both of these cotton fibers can be used as antibacterial products with a strength 4 to 5 times greater than in silver nanoparticles.

ACKNOWLEDGEMENT

The special thank delivered to the Agency of Research and Innovation National (BRIN) Republic Indonesia for financial support by Scheme of Applied Research 2020.

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[RJC-6034/2020]