



Effect Of Tetraethoxysilane Addition to Coconut Coir Ash Nanosilica on Morphology and Particle Size Of SiO₂-TiO₂ Nanocomposite

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Article History

Received : February, 16th 2024

Revised : February, 28th 2024

Accepted : March 31st, 2024

Published : March 31st, 2024

DOI:

<https://doi.org/10.24036/jeap.v2i1.48>

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Abstract: Nanotechnology is impossible without nano-sized materials. Nanomaterials can be used to produce composites that can be used in various fields, including in industry as a fabric coating material by providing certain properties to fabrics, one of which is water repellent properties. By adjusting the shape and size of the microscopic elements that make up the nanocomposite, these water-repellent properties can change the surface morphology, and interact with particles through the incorporation of two or more materials. This study aims to determine how the effect of tetraethoxysilane on Coconut Coir Ash Nanosilica on the Morphology and Particle Size of SiO₂-TiO₂ Nanocomposites. The method used in the extraction of coconut coir ash silica is the sol-gel method, which is very effective in producing nano-sized materials. After the nanocomposite was applied to the fabric, characterization was carried out with SEM and obtained particle sizes of 71 nm, 67 nm, 59 nm, 51 nm, respectively. The contact angles were 137°, 140°, 145°, 148°. The results of this study indicate that nanocomposites have better properties than their constituent particles. With the variation of adding 7 ml of tetraethoxysilane, the smallest particle size reaches 51 nm, and produces the best contact angle of 148°.

Keywords: Contact angle; Nanosilica; Particle size; Tetraethoxysilane; Titanium Dioxide.



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1. Introduction

Nanomaterials are nano-sized materials that are part of nanotechnology [1]. With more specific physical and chemical characteristics compared to larger materials, nanomaterials can be used to produce new materials that have better prospects for technological advancement [2]. The application of this technology is in water-repellent fabrics that are useful for maintaining the wettability of fabrics so that they can be used in several conditions [3]. Chemical composition and

How to cite:

T. D. Restika, Mardiana, S. F. M. Putri, Ratnawulan, Y. Darvina, and R. Hidayat, 2024, Effect Of Tetraethoxysilane Addition to Coconut Coir Ash Nanosilica on Morphology and Particle Size Of SiO₂-TiO₂ Nanocomposite, *Journal of Experimental and Applied Physics*, Vol.2, No.1, page 118-130.

environmental conditions usually determine material properties. A decrease in particle size and surface area increases the number of molecules or atoms on the surface, which can change the reactivity of the surface so as to make the fabric water repellent. Scientists are interested in nanomaterials, such as nanosilica and nanotitania that can be altered to create nanocomposite coatings that can be applied to fabrics [4].

The primary element in the earth's crust, silicon (Si), is a compound with the chemical formula SiO_2 that is found as nanosilica, or silicon dioxide. The industrial industry presently uses nanosilica, which is silica produced on a nano size (10^{-9} m). The product has unique qualities that might raise its quality because of the raw materials decreased particle size. The most common industrial application of silica is as the primary component of coatings for textiles and glass. Additionally, nanosilica is frequently utilized as a filler in product manufacture. Good stability, chemical inertness, biocompatibility that allows them to function in harmony with the body's system of functions, and single-crystal silica. Nanosilica can be found in various forms in nature, both in mineral and vegetable forms [5].

In vegetable form, silica can be obtained from coconut coir ash. A plentiful resource with significant pharmacological benefits, coconut coir is robust, lightweight, heat and salt resistant, weather resistant, inexpensive, and simple to access [6]. Coconut coir can be utilized as a raw material through the smelting process [7]. Coconut coir ash is the result of processing coconut coir waste that has been burned first using a certain temperature to produce ash. Potassium oxide (K_2O), magnesium oxide (MgO), zinc oxide (ZnO), iron oxide (Fe_2O_3), sodium oxide (Na_2O), silicon oxide (SiO_2) and phosphorus oxide (P_2O_5) are the chemicals found in coir ash [8].



Figure 1. Coconut Coir

The sol-gel method is one of many ways to get silica out of coconut coir ash [9]. The method uses organic silane as the precursor and hydrolyzes functional groups before condensing silane groups [10]. Naturally, this methods process is more intricate and involves parallel reactions such as the condensation of organic compounds from the precursor [11]. In the beginning, the hydroxyl group acts as a catalyst. It is essential to regulate the particle size, which has an average number at which degradation and condensing occur, by adjusting the pH level of the process. At high pH, condensation has a very fast rate compared to hydrolysis, fewer nuclei are formed and the reaction is easier to add monomer to the larger particles [12].

Another nano material is nano titania or titanium dioxide (TiO_2). The advantages of titanium dioxide semiconductors are that they have high photocatalytic activity, are stable, non-toxic, resistant to corrosion, and abundant, abundant, relatively cheap and has many applications [13]. TiO_2 has three phases crystals namely anatase, rutile and brookite [14]. Anatase and rutile which has a tetragonal structure has been widely studied for various applications photocatalysis.

Nanocomposites can be modified from the combination of 2 nanoparticles, namely the combination of SiO_2 and TiO_2 . This nanocomposite can create new functions that can be applied to various fields, one of which is as a coating material on fabric. However, in the synthesis process, the distribution of nanoparticles and the compatibility of SiO_2 fillers into the TiO_2 matrix are influenced by the amount of coupling agent added to the filler [15]. Coupling agents function as a bridge between the filler and the filler to form a strong bond between the surfaces of the two materials. In addition, the addition of coupling agents also affects particle size and surface morphology. Silane compounds are commonly used coupling agents [16]. Coupling agent is through chemical reaction and the surface of inorganic fillers accidentally coupling consolidation and crosslinked polymer binders, combine two kinds of different material, plays a role of bridge, from the bonding strength, raise yan, filler dispersion process in the basic material and reduce the extent of interfacial energy, coupling agent is greatly better than surfactant [17].

Biofunctional chemicals called silane coupling agents are frequently utilized to increase filler contact by chemical linkage, therefore increasing the effectiveness of silica reinforcement. Chemical reactions between two functionally active silane groups and silica and other matrices are possible [18]. On the silica surface, the hydrolyzable alkoxy group reacts with silanol to form a stable siloxane bond [19]. The interaction between silica and silane coupling agents has been the subject of several investigations, and a variety of silane coupling agent types have been produced commercially [20].

One of them is tetraethoxysilane (TEOS), a kind of low-volatile compound that is difficult to vaporize [21]. When in direct contact with water, tetraethoxysilane reacts quickly, becoming silicic acid and ethanol [22].

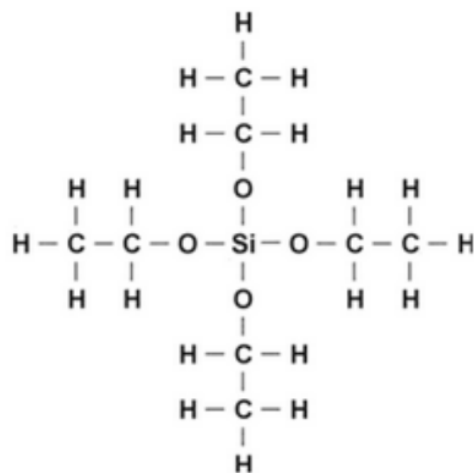


Figure 2. Tetraethoxysilane Structure

In addition, tetraethoxysilane is also insoluble in water, alkalis, mineral acids, and strong oxidizing agents. The form of tetraethoxysilane is a colorless liquid. Tetraethoxysilane includes silicone ester compounds, which are silicone compounds that have oxygen elements between silicones and are also organic groups such as $\equiv\text{SiOR}$ [23].

Another issue is that very few studies have shown how surface modification affects dispersion stability and particle interactions with silane coupling agents. This is due to the fact that achieving homogenous polymer composites and preventing the development of aggregates throughout the surface alteration process require careful management of the dispersion behavior and interactions of particles in organic solvents. Thus, in order to find out how silica functions to improve its capacity to interact with other matrices, tetraethoxysilane-type silane bonding substances were employed in the study. Previous research by [24] only used two materials, namely SiO_2 and TiO_2 , the novelty of this research is the surface modification with silane compounds, namely tetraethoxysilane, where the addition of this modification will strengthen the bond between the two nanocomposites so as to produce a better coating.

2. Materials and Method

This category of study is known as experimental research. This study investigates the effects of tetraethoxysilane on the morphology and particle size of cotton fabric when added to coconut coir ash nanosilica. The tools that have been used in this experiment include measuring cups, beakers, Erlenmeyer, oven, magnetic stirrer, measuring pipette, dropper pipette, digital scales, and DSLR camera. While the characterization tool that has been used is SEM. The materials used in this research are coconut coir ash, titanium dioxide, tetraethoxysilane, sodium hydroxide (NaOH), chloride solution (HCl), distilled water, alcohol, and xylene.

In this research there are several processes that must be carried out as shown in Figure 3.

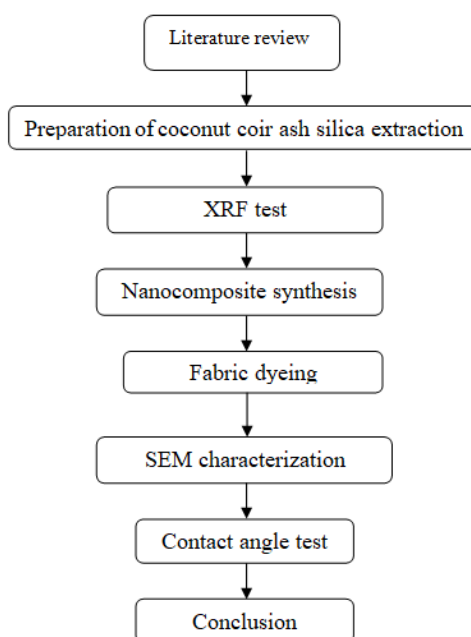


Figure 3. Research Design

The following steps are taken in making the SiO₂/TiO₂ nanocomposite layer: cutting coconut fiber into smaller sizes. Then clean the coconut fiber first so that the coconut fiber is free from impurities that can affect silica yield. The coconut fiber is then cut into smaller sizes. After that, it was oven-dried and fumigated at 700°C. The ash was then pulverized using High Energy Milling for 6 hours. Next, the coconut fiber ash was added to 0.1 M NaOH solution and placed on a magnetic stirrer. Stirring was carried out continuously at 50°C for 1.5 hours. Then the resulting solution was filtered with Na₂SiO₃ to separate the precipitate. The Na₂SiO₃ solution was added gradually to 0.1 M HCl until the color of the solution changed slightly, then tetraethoxysilane solution was added until a white gel precipitate was produced, to obtain the maturity of the gel, the gel was allowed to stand for 48 hours. After that, the gel was washed until the washing water was white and then dried in an oven to dry.

The SiO₂/TiO₂ composite synthesis stage was carried out by the sol-gel method. First, weigh 3 grams of TiO₂ and mix it with 30 ml of toluene. In a separate container mix 2 grams of SiO₂ with 30 ml of toluene. Then mix the two solutions and stir for 1 hour. Then let stand for 24 hours until a homogeneous solution is obtained. The cotton fabric coating stage is carried out by preparing a 3 x 3 cm cotton cloth and sterilizing it first using an ultrasonic cleaner. Then the cotton cloth was soaked with alcohol in a 100 ml beaker for 5 minutes in an ultrasonic cleaner. Next, the cotton fabric was removed and dried using an oven for 5 minutes at 80°C so that the cotton fabric was ready to be coated. The fabric coating is done by dipping the fabric into the SiO₂/TiO₂ composite solution for about 5 minutes then removed using tweezers and placed in a closed room and waited to dry. The required drying time is 5 minutes in the oven at 90°C. Then characterization is carried out using SEM to see the morphological structure of the fabric that has been coated.

3. Results and Discussion

According to the results of the silica synthesis that was completed. Additionally, X-Ray Fluorescence (XRF) testing was done to ascertain the elemental content and chemical composition of coconut coir ash following extraction, the findings are displayed Table 1.

Table 1. Element content of XRF test results

Elements	Percent composition
SiO ₂	94,766%
P ₂ O ₅	3,171 %
CaO	0,598 %
Ag	0,319 %
Cl	0,005 %
Ti	0,146 %
Fe ₂ O ₃	0,088 %
Zr	0,004 %
Ba	0,004 %
Al ₂ O ₃	0,879 %

Based on the table of XRF test results after extraction or purification, the highest silica content is 94.7% in the addition of 7 ml tetraethoxysilane. With the composition of other impurities with

the most percentage is P_2O_5 with a percentage of 3,171 %, so it can be considered that the silica purification is quite high, even though there are still many impurities. This is because the purification process is not perfect, so there are still other materials. The silica recovered from acid treatment was found between 25% to 68% while alkali treatment was at 76% [25]. That means the more addition of tetraethoxysilane will increase the silica content because the addition of tetraethoxysilane in the fixed volume silylation process can replace the silanol-bound OH group with alkoxy silica. Silica also plays a role in improving surface roughness with the addition of co-precursors such as tetraethoxysilane.

Research by [26] also demonstrated a different approach to producing silica from leftover agricultural products at a lower manufacturing cost, effectively synthesizing SiO_2 from coconut coir ash. The proportion of silica that was recovered efficiently raised from 9.24% to 91.76% using chemical treatment [27]. It is demonstrated that the structure and morphology of silica change throughout the extraction process, resulting in silica in crystalline form when acid-catalyzed with an amorphous phase. As coir ash has a high silica purity, it might be a viable substitute supply of silica to lower manufacturing costs.

Morphology test or SEM test (Scanning Electron Microscopy) nanocomposite SiO_2/TiO_2 conducted to determine the distribution of SiO_2 filler modified tetraethoxysilane (TEOS) in the TiO_2 matrix which affects the morphology and particle size. SEM (Scanning Electron Microscopy) results in the form of filler distribution images with various magnification scales. Scale magnification scale that will be used for This research is 2000 times magnification. The samples tested by SEM are fabric samples that have been successfully coated with nanocomposites by varying tetraethoxysilane (TEOS), namely 1 ml, 3 ml, 5 ml, and 7 ml. The fabric without coated SiO_2/TiO_2 nanocomposites can be observed in the Figure 4.

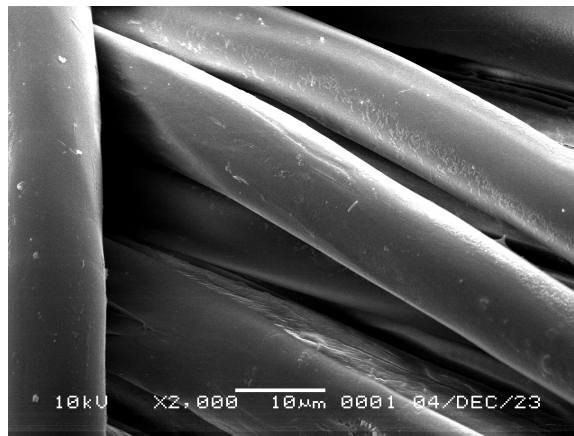


Figure 4. The Morphology of Cotton Fabric Without Coating

It is clearly seen from Figure 4 that the cotton fabric fibers have a clean surface morphology and are not coated by the SiO_2/TiO_2 nanocomposite layer. The difference in fabric surface morphology after coating with SiO_2/TiO_2 nanocomposites with variations in the addition of tetraethoxysilane can be observed in the Figure 5.

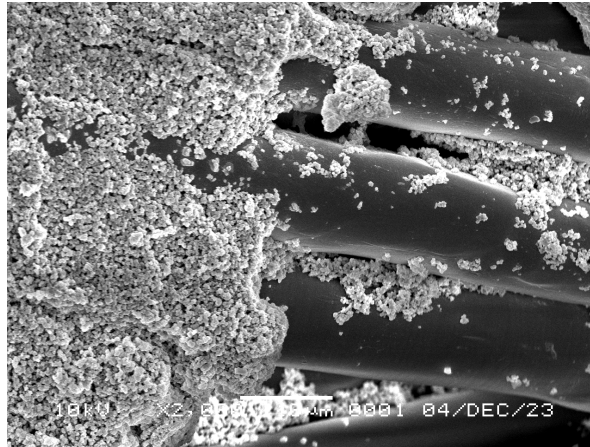


Figure 5. Fabric morphology with 1 ml *TEOS* variation

The difference between fabrics that are not coated with nanocomposites and after being coated with nanocomposites is on the surface of the fabric which looks like there are layers or nanoparticles attached to cover the fabric fibers as seen in Figure 5. For the variation of 1 ml tetraethoxysilane addition can be seen in Figure 5. The resulting particle structure is classified as heterogeneous. In the variation of adding 1 ml tetraethoxysilane, it can be seen that there are still many fabric fibers that have not been coated with nanocomposites, this is likely due to the combination of particles in the nanocomposite so that the SiO_2 particles are not well distributed in the matrix and make the nanocomposite unable to stick firmly to the fabric, although some are attached but unevenly. The use of coupling agents has not produced optimal morphology. Where the distribution and interaction of filler and matrix are not homogeneous.

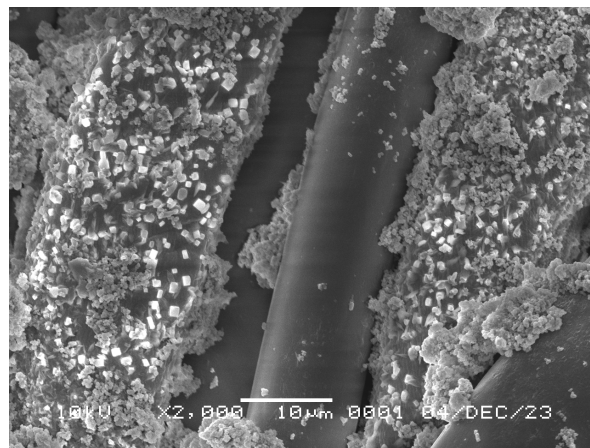


Figure 6. Fabric morphology with 3 ml *TEOS* variation

According to Figure 6, In the addition of 3 ml tetraethoxysilane the results are almost the same as 1 ml tetraethoxysilane where the resulting particle structure is still classified as heterogeneous. And there are still parts of the fabric fiber that are not coated with nanocomposites, this can also be caused by a less than optimal coating process so that nanocomposites cannot stick to the fabric surface.

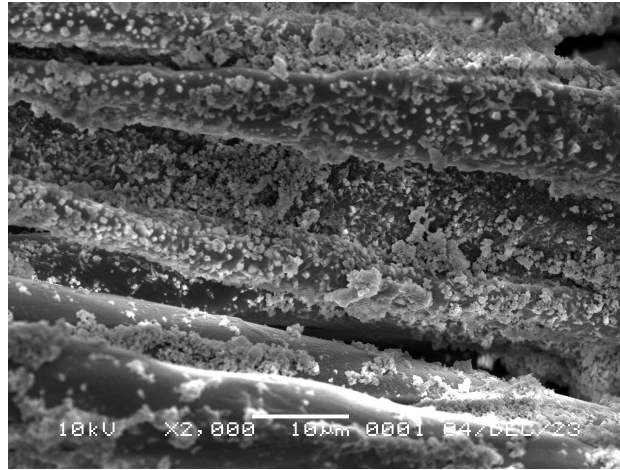


Figure 7. Fabric morphology with 5 ml *TEOS* variation

The addition of 5 ml tetraethoxysilane shows a better structure than the previous variation, where almost the entire surface of the fabric is coated with nanocomposites with a blob-shaped structure with a few protrusions, indicating that the silane coupling agent (tetraethoxysilane) starts to work well so that it can produce a homogeneous structure. Along with the addition of tetraethoxysilane shows the results of the morphological structure that is getting better, for the addition of 7 ml tetraethoxysilane observable in the Figure 8.

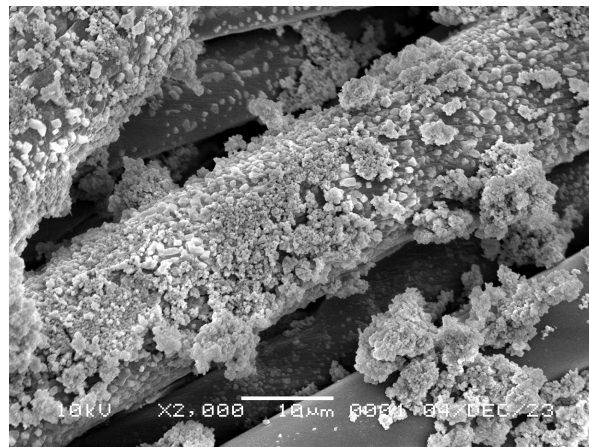


Figure 7. Fabric morphology with 7 ml *TEOS* variation

The best result is the addition of 7 ml tetraethoxysilane shows that almost the entire surface of the fabric is successfully coated with nanocomposites and produces clumps in the form of bulges. According to [28] Because many protrusions are produced, each particle has a high roughness. The incorporation of particle bulges will increase the bonding between particles on the fabric surface. In accordance with research showing that the MNR coupling agent works well at the highest level, namely MNR 3 phr, the coupling agents polar groups will attach to the fillers polar groups, while its non-polar groups will attach to the matrix's non-polar groups.

Table 2. Particle size data

TEOS Variation (ml)	Particle Size(nm)
1	71
3	67
5	59
7	51

The average particle size of each sample can be seen in Table 2. Based on the calculation of the SEM results, it can be concluded that the addition of tetraethoxysilane causes a decrease in the particle size of the nanocomposite, but forms clumps between particles (agglomeration). Smaller particle size will increase the surface area of contact between particles with liquid, as a result it will increase the mass transfer besides it will also minimize the diffusion distance [29]. The cause of this decrease in particle size is because the hydrophilic group of SiO₂ filler interacts with coupling agents that contain hydroxyl groups and at each additional concentration of tetraethoxysilane concentration will increase the formation of micelles. Micelles are aggregates of amphipathic molecules in water with the nonpolar being on the inside and the polar part is on the outside exposed to air. This suggests that adding a silane coupling agent is a highly recommended action, considering that the hydroxyl group of silane can bind to the silica and is able to lower the surface tension, as well as making the particle size smaller than before and prevent coagulation. Therefore, it can be identified that the addition of tetraethoxysilane greatly affects the particle size of the nanocomposite.

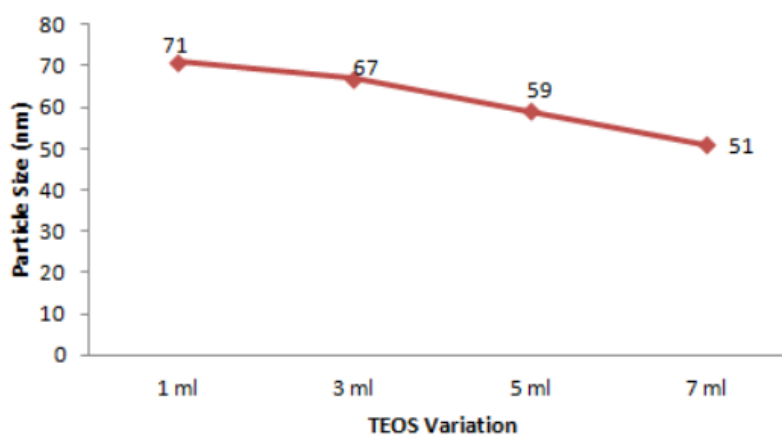


Figure 9. Graph of tetraethoxysilane effect on particle size

Based on Figure 9, it can be seen the effect of the addition of tetraethoxysilane on the size of the nanocomposite, the addition of tetraethoxysilane produces a small particle size. Due to the addition of tetraethoxysilane has worked optimally as a SiO₂ reinforcement that contributes to increasing the surface area by reducing the particle size around TiO₂ particles so that the adsorption side is getting bigger. In research by [30], silane-modified nanocomposites influence the size of composite nanoparticles; the most efficient modification was obtained VS, having a mean size of the particles of 7.8 ± 1.0 nm and a distribution of particle sizes of 83% (≤ 10 nm). In accordance with research [31] the inclusion of silane coupling has an impact on the size of the particles; the smallest particles generated, when utilizing hexyltrimethoxysilane as a coupled agent, are 30 nm in size. The structure of the surface of silanols and the way they react with C6S are significantly influenced by the size of silica particles..

To observe the effect of particle size on the surface properties of the fabric, contact angle testing can be done using a thermal camera. The following is an image of the contact angle measurement results based on the variation of tetraethoxysilane addition presented in Figure 10.

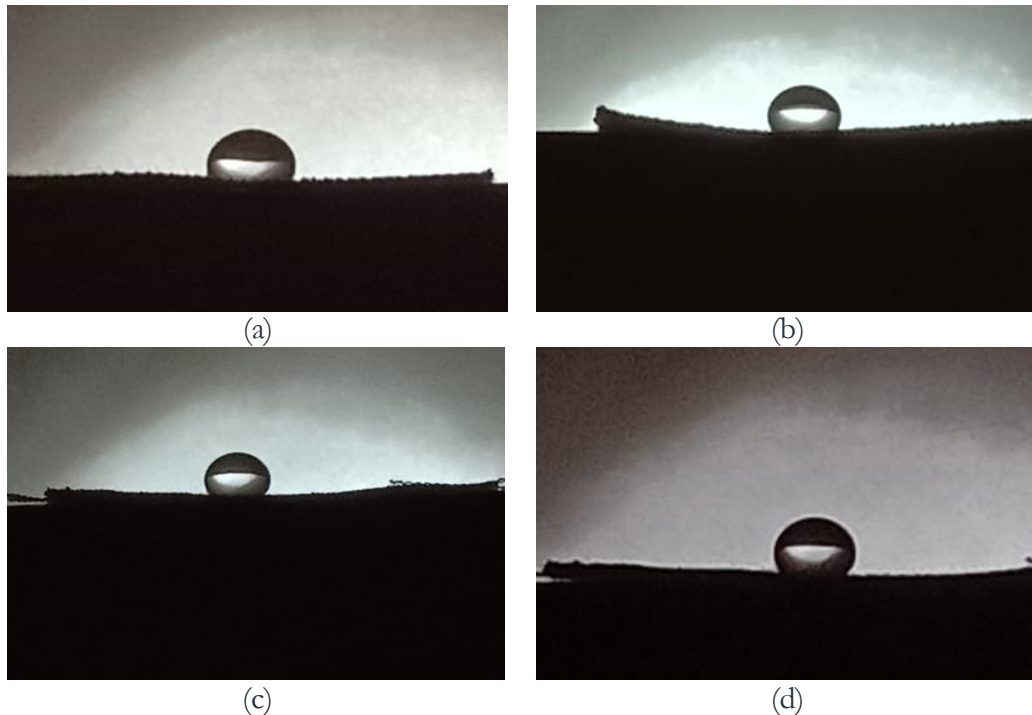


Figure 10. Results of contact angle of nanocomposite coated fabric (a) 1 ml TEOS variation (b) 3 ml TEOS variation (c) 5 ml TEOS variation (d) 7 ml TEOS variation

Figure 10 shows the contact angle of each fabric variation. By using Image-J software, the contact angle formed can be calculated. The test results with Image-J software can be observed in the Table 3.

TEOS Variation	Contact angle
1 ml	137°
3 ml	140°
5 ml	145°
7 ml	148°

Based on the test results and measurements of the contact angle between water and the surface of the coating, it can be seen that the addition of tetraethoxysilane to the silica filler has a major effect on the contact angle formed between the four mass variations. The four versions are water-repellent and have been created successfully. The highest contact angle is formed in the variation of adding 7 ml tetraethoxysilane, which is 148°. Meanwhile, the variation of adding 1 ml tetraethoxysilane is 145°. The 3 ml and 5 ml addition variations were seen to have a fairly large increase in contact angle. All contact angles produced can make the fabric water repellent because the contact angle is in the range of $>90^\circ$ and is included in the hydrophobic layer.

This is also supported by the size of the nanocomposite particles attached to the fabric which is quite small at 51 nm and its rough surface. This study shows results towards smaller particle sizes

as the addition of tetraethoxysilane increases. This contributes to producing a rougher surface that makes the surface hydrophobic. Previous research by [32] which also used SiO₂/TiO₂ nanocomposites but without using tetraethoxysilane produced an average particle size of 202.98 nm with the highest contact angle of 110°. From this research, it can be identified that nanocomposites without the addition of coupling agents produce nanocomposites with particle sizes that are not yet nano, so it can be identified that the addition of tetraethoxysilane is very important in order to get nano-sized particles so that it contributes to increasing the contact angle.

4. Conclusion

The manufacture of nanocomposites by utilizing nanomaterials has been successfully carried out. The addition of 7 ml tetraethoxysilane obtained the smallest particle size of 51 nm. This particle size has an impact on the morphological roughness of the nanocomposite coating, because nanocomposite materials have stronger chemical properties and lower surface energy if the particle size is smaller. The maximum contact angle, which is 148° degrees, is attained in a variance of 7 ml tetraethoxysilane in order to improve the fabric's surface contact angle. The nanocomposite layer formed has the best interface morphology and compatibility between TiO₂ matrix and silane-modified nanosilica filler type tetraethoxysilane.

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