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Physical Strength Improvement of Nata de Coco by Water Replacement with Carboxymethylcellulose (CMC) as A Potential Bulletproof Material: A Preliminary Study

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Abstract—The dependence on the imported bullet-proof vest as one of the main equipments of the National Defense System needs attention. This condition treats the independence of domestic defense security. In contrast, the potential of natural materials for bullet-proof vest plates are abundant in Indonesia. Nata de Coco is one of the natural raw materials for producing bullet-proof vest plates that has the potential to be developed. This preliminary research proved that increasing the bond strength of cellulose in Nata de Coco was performed by adding the appropriate crosslinkers. The addition of carboxymethylcellulose (CMC) 2% crosslinker to form Nata de Coco kevlar-like plates (Navlar) was evidenced to increase threefold tensile strength (99.15 MPa) from its original Nata de Coco (31.92 MPa). The development of Navlar is a very strategic sector for producing high-quality bullet-proof vest plates equivalent to kevlar strength. Navlar is more prospective than Kevlar due to its abundant source, cheap, lightweight, and ease to manufacture. Developing Navlar could replace the dependence on imported Kevlar and support the domestic defense industry.

Keywords— Bulletproof material; CMC; Kevlar plate; Nata de Coco.

1. INTRODUCTION

As one of the main defense equipment, the bullet-resistant vest development is essential. Hence, the necessity and independence of domestic plate production should be the main priority for the defense industry. Generally, the plates are made of poly-aramid polymer known as Kevlar [1].

The plate made of kevlar is widely used by soldiers as a shielding component of body armor from bullet attacks in military operation conditions involving the use of weapons. Based on data from The International Institute for Strategic Studies (IISS), Indonesia ranks 8th based on the number of active soldiers, which is around 438,410 personnel [2]. Compared to the population ratio, the number of Indonesian soldiers is still relatively low. Compared to other countries in the world, Indonesia's military strength ranks 14th (**Table 1**).

Considering the statistics of Indonesian military personnel, which need to be improved and enhanced every year, is aligned with the domestic plate production. This becomes one of the main concerns of the Republic of Indonesia Ministry of Defense in an-

Table 1. Map Military Strength and Reserves of Countries in the World [2]

Rank	Country	Total Population (thousand)	Active Soldier (thousand)	Reserve Army (thousand)
1	China	1,405,930	2,255	800
2	America	334,297	2,580	2,459
3	Russia	146,877	2,037	2,400
4	North Korea	25,491	1,106	3,200
5	South Korea	51,446	687	1,500
6	India	1,366,650	1,414	1,115
7	Pakistan	221,278	700	528
8	Vietnamese	93,700	348	4,000
9	Singapore	5,612	61	313
10	Thailand	69,038	310	245
11	Malaysia	32,570	110	42
12	Philippines	111,235	106	131
13	Timor Leste	1,319	2	-
14	Indonesia	268,583	439	439

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attempt to strengthen one of the modern and reliable elements of national defense equipment.

Based on its specifications as an element of defense equipment, the plate in a bullet-proof vest consists of several types, including soft body armor and hard body armor [3]. Soft body armor is a bullet-proof plate with less than 1 mm thickness, consisting of 32 layers weighing up to 10 kg. This type of bullet-proof plate tends to be lighter so it is often used in undercover duties and as a security component for soldiers personnel. Meanwhile, hard body armor is a plate made of ceramic material composited with a certain metal plate. This makes hard body armor tend to be heavier and thicker so that it is used only for special tasks that have a high risk to the safety of soldiers [4].

Until now, the Indonesian defense industry has still been unable to produce bulletproof plates independently. Kevlar plate production in Indonesia is also still considered too expensive because it requires expensive raw materials and infrastructure. Commercial kevlar is synthesized in a solution from the monomer 1,4-phenylenediamine (para-phenylenediamine) and terephthaloyl chloride in a condensation reaction which produces hydrochloric acid as a side product [5]. Then, this reaction also produces a polymer in liquid crystal form. The formation reaction of Kevlar was first discovered by American chemist Stephanie Kwolek while working for DuPont. Stephani's work is patented under the Kevlar brand as Poly-paraphenylene terephthalamide (K29). The reaction equation for the formation of Kevlar is presented in **Fig. 1.**

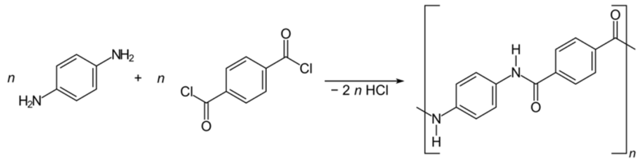

Fig. 1. Commercial Kevlar formation reaction

Previous research conducted by the Indonesian Science Institute (LIPI) shows that hemp has a modulus of elasticity equivalent to Kevlar. The modulus elasticity of hemp is around 44-90 gigapascals, whereas Kevlar is around 40-140 gigapascals [6]. However, the breaking strain in hemp is higher than in Kevlar (2% hemp and 1-3% Kevlar). The density of hemp and Kevlar is almost the same. Hemp is 1.50 grams per cubic centimeter and Kevlar is 1.45 grams [7]. Furthermore, the Head of the Polymer Testing Laboratory, Rahmat Santoto, claimed that the research on the hemp plant is finally stopped because the material from hemp fiber is considered uneconomical to be used as bullet-proof clothing [7].

In addition to the reinforcing materials, another item that needs to be considered is the matrix. The appropriate matrix will have a good impact on the mechanical properties and microstructure of the composite. A material that can be used as a matrix is epoxy containing an oxirene group structure [7–10]. The epoxy resin matrix has better corrosion resistance than polyester in wet conditions and has a thick or almost solid-liquid appearance. In addition, epoxy has better mechanical, electrical, dimensional stability, and heat retaining properties [11]. This resin is commonly combined with a hardener which then forms a crosslink polymer at room temperature. The type of hardeners for curing systems are generally polyamide compounds consisting of two or more amine groups [12].

To fulfill the need for bullet-resistant materials that are lighter, cheaper, and the raw materials are available domestically, research focusing on bullet-resistant materials is very strategic and essential to perform. One candidate polymer material that has properties similar to the constituents of Kevlar is Nata de Coco [13]. Nata de Coco, made from coconut water, is an abundantly available ingredient in Indonesia. Nata de coco consists of cellulose units with a high degree of crystallinity. Nata de Coco, consisting of layers of cellulose and amylose (starch), is a polymer that is distinguished by the type of glycosidic bond. Cellulose has an alpha-type glycosidic bond, while amylose has a beta-type (**Fig. 2**).

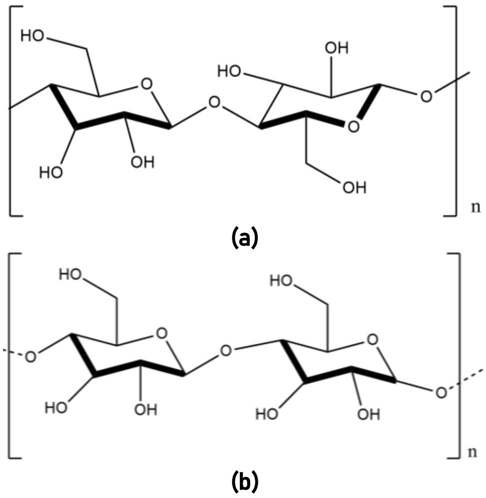

Fig. 2. Glycosidic bonds in (a) cellulose and (b) amylose

The latest research on Nata de Coco as a bulletproof material candidate is conducted by Kristanti et al. who composites Nata de Coco with hemp fiber [13]. Unfortunately, the results shows that at 6 layers Nata de Coco can still be penetrated by a 9 mm caliber revolver at a distance of 10 m. The arrangement of the carbon chains that make up Nata de Coco cellulose is the main key to the strength of natural fibers. However, in the form of natural fibers, its strength does not depend entirely on the cellulose content but on controlling fiber strength recisely from the holes or fractures that occur in the fiber [12,14]. The fiber strength also depends on the proper crosslinker so that the fibers can compact and coalesce.

Based on the literature study conducted in this research, the physical modification of Nata de Coco has been carried out as the basic material for making bullet-resistant materials. However, the modification of the crosslinker to bind cellulose fibers in Nata de Coco has never been carried out. This research contains a preliminary study on the effect of carboxymethylcellulose (CMC) as the crosslinker on the physical properties of Nata de Coco as a candidate for bullet-resistant plates. The authors hopes that this research can be continued until it becomes a Navlar product that uses cheap and easy-to-obtain raw materials but produces products that meet military standards.

2. EXPERIMENTAL SECTION

2.1. Materials

Samples of Nata de Coco was provided by CV. Agrindo Suprafood located in Bantul, Yogyakarta. The citric acid monohydrate and CMC with moleculer weight ~90.000 were analytical grade provided by Merck® and Sigma Aldrich, respectively.

2.2. Instrumentations

The equipment used in this study included an oven, self designed grinder and miller, and the analytical balance (Mettler AE100). Functional group analysis was conducted by FTIR (Shimadzu Prestige 21). Physical strength measurement was performed by universal testing machine (UTM) Zwick 0.5.

2.3. Navlar Manufacturing Process

A preliminary study was conducted by applying a chemical treatment of Nata de Coco, which would be used as Navlar. Firstly, the wet Nata de Coco was cut into small pieces of 10 × 10 cm. Then, the small pieces of Nata de Coco were treated according to the treatment shown in **Table 2.**

Table 2. Preliminary study treatment toward the effect of CMC crosslinker and the number of Nata de Coco layers

Number	Code	Treatment	Solution Information
1	N-01	Nata de Coco non-milled	Soaking in 2% citric acid, RTP, 24 h.
2	N-02	Nata de Coco milled	Soaking in 2% citric acid, RTP, 24 h.
3	N-03	Minced Nata de Coco + CMC 2% - 1 layer	Soaking in 2% citric acid, RTP, 24 h.
4	N-04	Nata de Coco ground + CMC 2% - 2 layers	Soaking in 2% citric acid, RTP, 24 h.
5	N-05	Nata de Coco ground + CMC 2% - 3 layers	Soaking in 2% citric acid, RTP, 24 h.

After the preparation process, all samples N-01 to N-05 were soaked for 24 h in a solution according to the treatment in **Table 2**. The samples were then dried in an oven at 60 °C. Subsequently, a Fourier Transform Infra-Red (FTIR) spectroscopy, the tensile strength, and elasticity tests were carried out to determine the chemical bond changes in the sample, and the physical properties of the sample, respectively.

3. RESULT AND DISCUSSION

3.1. Appearance of Navlar

The Nata de Coco grounding process before soaking is intended to remove the water contained in the Nata de Coco, it can also support CMC and citric acid to enter into the Nata de Coco cellulose fibers and make them stronger. The process of cutting, grounding, and soaking Nata de Coco is presented in **Fig. 3.**

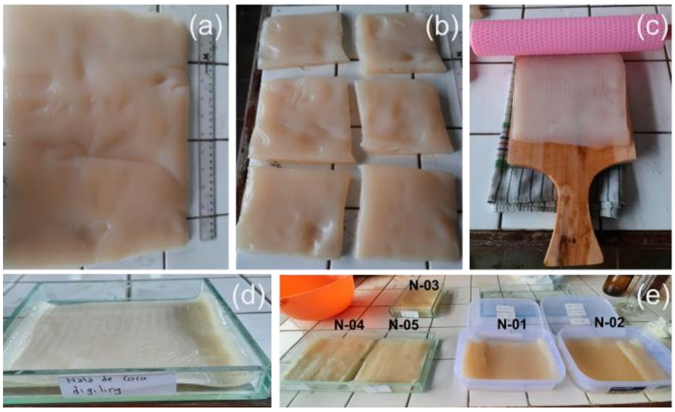

Fig. 3. (a) Nata de Coco sheets; (b) Nata de Coco cut into 10×10 cm size; (c) Nata de Coco milling process; (d) Appearance of Nata de Coco after grinding; and (e) Sample preparation with codes N-01 to N-05 has been completed and the immersion process will continue.

Fig. 4. (a) Sample immersion process; (b) Appearance of the sample after being baked; (c) Appearance of the whole sample; and (d) samples ready for characterization.

After the drying process, the color of the sample was brown with a skin-like texture (**Fig. 4b**). Visually, the sample was very ductile and quite elastic. The sample appearance in the soaking process can be seen in **Fig. 4a**. The different colors between the nata de coco sample and the matrix was showed in **Fig. 4c**. The dark brown of the sample shows the sample and matrix appearance after the drying process and the light brown color of the sample in **Fig. 4c** is the sample also the matrix appearance without the drying process. The matrix sample in **Fig. 4d** showed the appearance of N-01 to N-05 that were ready for characterization.

3.2. Physical and chemical properties of Navlar

The samples of Navlar with the codes N-01 to N-05 were then tested for tensile strength and elongation to observe the resistance and effect of treatment on each sample. The physical test was conducted duplo. The results are displayed in **Table 3.**

Table 3. Navlar tensile strength and elongation test results with codes N-01 to N-05

Number	Code	Tensile Strength (MPa)	Elongation (%)
1	N-01	31.92	12.19
		35.00	11.11
2	N-02	28.41	16.24
		27.34	14.90
3	N-03	81.75	5.63
		71.89	7.12
4	N-04	91.60	7.05
		78.63	7.84
5	N-05	99.18	7.18
		93.15	5.45

There was a significant difference between ungrounded (N-01) and grounded (N-02) Nata de Coco. It can be known that the ungrounded Nata de Coco has weaker physical properties than the grounded Nata de Coco because it still contains some water. However, compared to the sample N-02, the addition of CMC in samples N-03 to N-05 increased the tensile strength up to about 300%. Comparing this result with Kevlar, which had 3500 MPa tensile strength, N-05, which had the highest tensile strength, was still not close.

This result opens insight into the significant impact of crosslinker use on the strength enhancement of Nata de Coco. The crosslinker used in this study CMC as a cross-linking agent that has a low-degree of substitution and often used as an additional component in industrial materials [15]. CMC has a significant role in exhibiting the water content up to 94% and enhance the compressive strength up to 80 MPa [16]. The usage of CMC can produce significant yield strength by creating stong-covalent bonds between the composite structuress [17].

The samples were analyzed using FT-IR to explain the effect of CMC on the physical strength of the sample. There was no significant differences between functional group content of CMC and Navlar (**Fig. 5**). However, the fine increases was obtained in the intensity of the number of C–O bonds in Navlar indicated the cellulose functional group in Nata de Coco at wave numbers 1050–1085 cm^{-1} (**Fig. 5**). From these results, it can be predicted that if we want other Kevlar-like bonds, like C-N (amide) then the crosslinker only needs to be varied with nitrogen-containing groups, such as cyanonitrile, acetonitrile, and so forth.

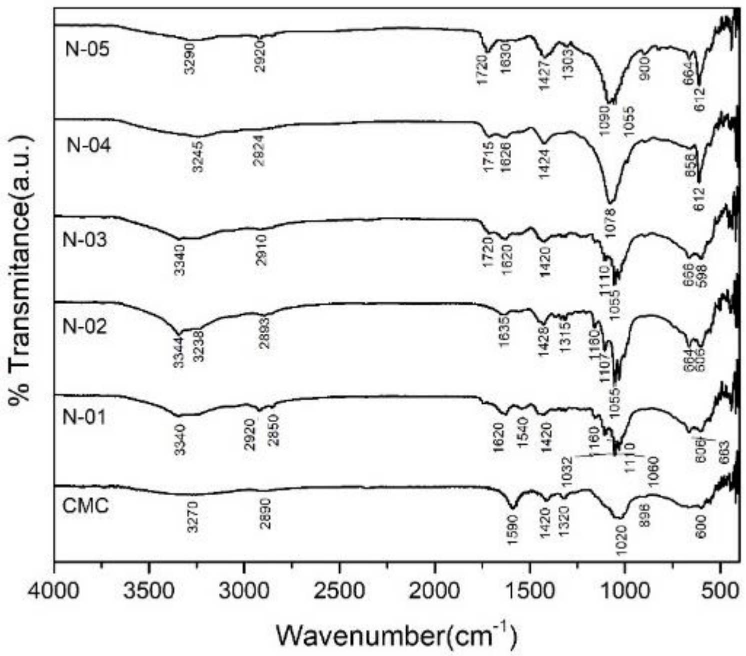

Fig. 5. FT-IR spectra of CMC, N-01, N-02, N-03, N-04, and N-05

4. CONCLUSION

The utilization of Nata de Coco as a raw material for bullet-resistant materials is very potential to be developed. This preliminary research proved that the increase in the bond strength of cellulose in Nata de Coco could be achieved by adding a proper and suitable crosslinker. The addition of a 2% CMC crosslinker to Nata de Coco could increase the tensile strength of the N-05 sample by 300%.

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CONFLICT OF INTEREST

The authors have no conflict of interest in this paper.

AUTHOR CONTRIBUTIONS

BI, PIWB, and RB conducted the experiment; AS, DCT, and FR wrote and revised the manuscript under supervision of RB and MF. All authors agreed to the final version of this manuscript.

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