

# Synthesis and characterization of hydrogel composite based on bacterial cellulose-gambir leaf extract (*Uncaria gambir* Roxb.)

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## Synthesis and characterization of hydrogel composite based on bacterial cellulose-gambir leaf extract (*Uncaria gambir* Roxb.)

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**Abstract.** Synthesis and characterization of hydrogel composite based Bacterial Cellulose - Gambir Leaf Extract (BC-GLE) has been done. The purpose of this research was to obtain an elastic hydrogel composite BC-GLE that can be applied as a substitution material for cartilage. Preparation of this hydrogel composites conducted by immersing BC in GLE for 1, 2, 3 and 4 days. The effects of immersion time on the physical and mechanical properties of BC-GLE were studied by measuring its water content, compressive and tensile strength. The structures of BC-GLE were analyzed by using FTIR and XRD. The results showed an improvement of physical and mechanical properties of BC-GLE hydrogel composites in line with the duration of immersion. Absorption of GLE into BC matrix caused decreasing the water content of BC. The best water content percentage result was obtained from the BC-GLE composite which was immersed for 4 days, 98.211%. The best compressive strength result was also obtained from samples on the immersion time of 4 days, 2.633 MPa. The tensile strength of the BC-GLE hydrogel composite increased from day 1 to 3, but decreased in day 4. The best tensile strength result of 0.17 MPa was obtained on the day 3 immersion time. Physical and mechanical properties improvements of BC-GLE hydrogel composite were only caused by absorption of GLE to BC matrix without forming new bond. The FTIR results showed no new functional group was formed, but only showed vibration shift to a lower wave number. BC-GLE hydrogel composite structure was still in a cellulose type 1. Combination of BC and GLE produced BC-GLE hydrogel composite material that having physical and mechanical properties better than pure BC. However these properties values were not reached the standard to be applied as a substitute material for cartilage yet.

### 1. Introduction

Design and fabrication of a suitable material for soft tissue substitutes is an important aspect for biomedical applications. This encourages the need to develop a material that not only exhibits the same mechanical properties as the soft tissue to be replaced, but also exhibits an improvement of functionality, biocompatibility and durability. A material that is promising and appropriate for this purpose is hydrogel. Hydrogel is a network of three-dimensional hydrophilic polymers formed from the reaction of one or more monomers or by the incorporation of bonds between polymer chains. Hydrogel can absorb water from 20% to thousands of times its dry weight. This unique property may



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cause hydrogel to be used in various medical, pharmaceutical and prosthetic applications such as contact lens, synthetic wound cover, drug delivery system, organs, and soft tissue [1].

Bacterial cellulose (BC) is one of the hydrogels produced by bacteria from the genus *Acetobacter* such as *Acetobacter xylinum*. BC has many advantages such as high purity, excellent network structures, unique mechanical strength and high water content (98-99%), and can be sterilized safely without changing its characteristics [2]. BC has high tensile strength along the fiber layer direction, but has low compressive modulus value. Therefore, BC is less elastic [3]. Utilization of BC in the biomedical field as a substitute material for soft tissue is constrained by BC's low elasticity properties. To improve the BC elasticity, it is necessary to combine BC with other materials that produce a composite material with better physical and mechanical characteristics [1]. One alternative way to produce a good composite material is to combine BC with natural materials such as gambir leaf extract.

Gambir plants (*Uncaria gambir* Roxb.) are herbs that are widely found in Indonesia. About 90% of Indonesia's gambir production is produced in West Sumatra. Gambir plants contain compounds that are high in antioxidants that have been exploited in the field of pharmaceuticals as anti-inflammatory drugs, astringent, and even potentially anti-cancer drugs [4] [5]. Gambir leaf is commonly used by people to treat various diseases such as dysentery, diarrhea, and canker sores. The main content of gambir leaf extract (GLE) is Catechin compound (7-33%), catechu tannat acid (20-55%), Pyrocatechol (20-30%) and quersetin (2-4%) [7].

Based on the above description, a research was done by combining BC with GLE which is expected to produce a material of Bacterial Cellulose-Gambir Leaf Extract (BC-GLE) which can be applied in the biomedical field.

## 2. Method

### 2.1. Materials

The equipment used were glassware (pyrex), stainless steel container, plastic container, mixer/blender, vapor plate, plastic, iron (Sanyo), analytical balance (KERN ABS 220-4), oven (Mettler Model 300), filter, thermometer, pH meter (Seven Easy S20 METTLER-TOLEDO), FTIR (PerkinElmer Frontier Optica), and Compressive tester (Tony Technik Compressive Test Bauform model 2020). The material used in this research were waste coconut water,  $\text{CO}(\text{NH}_2)_2$  (PT. Pupuk Sriwijaya),  $\text{C}_{12}\text{H}_{22}\text{O}_{11}$ ,  $\text{CH}_3\text{COOH}$  25% (PT. Brataco Chemica), starter of the *A. xylinum* obtained from Biochemistry Laboratory of Universitas Negeri Padang, aquades, and gambir leaves was obtained from Siguntur Gambir plantation, Pesisir Selatan, West Sumatera, Indonesia

### 2.2. BC fermentation

Four liters of filtered waste coconut water was poured into a 5 L stainless steel container, then heated and added 400 g of  $\text{C}_{12}\text{H}_{22}\text{O}_{11}$ , 40 g of  $\text{CO}(\text{NH}_2)_2$ , and stirred. Then, the mixture was acidified with 25% of  $\text{CH}_3\text{COOH}$  to reach pH 4-4.5 ( $\pm 80$  mL), and then heated. In hot conditions, 600 mL of the mixture was poured into a sterilized fermented container and closed using sterile pore paper. The mixture was cooled to a temperature of  $\pm 28^\circ\text{C}$  (room temperature), and then aseptically added 10% v/v (60 mL) *A. xylinum*. This mixture was fermented for several days to form BC with a thickness of  $\pm 1$  cm.

### 2.3. Washing and purifying BC

The obtained BC was washed with running water for  $\pm 24$  hours, and then was immersed in NaOH 2% (w/v) solution for  $\pm 24$  hours to remove impurities and bacterial cells. The pure BC was then washed again by using running water for  $\pm 24$  hours for further storage, and ready to use.

#### 2.4. Gambir Leaf Extract preparation

The clean old gambir leaves were cut into small pieces and mashed with a blender. Then, the leaves were soaked for  $\pm 15$  minutes in hot water (temperature  $\pm 90^\circ\text{C}$ ) with a ratio of leaf and water of 1: 4. The mixture was stirred and filtered until a brownish extract was obtained. This brownish extract was used for the preparation of BC-GLE hydrogel composite.

#### 2.5. BC-GLE hydrogel composite preparation

BC was cut in size of 2x2x1 cm and 15x2x1 cm, and inserted into a plastic container. Into this plastic container, 500 mL GLE was added and placed on the shaker. BC in GLE was shaking for 1, 2, 3 and 4 days. Each day, the resulting BC-GLE hydrogel composite was removed from the plastic container and the sides of the surfaces were dried by using tissues. The BC-GLE samples were then used for characterization

#### 2.6. BC-GLE hydrogel composite characterizations

##### 2.6.1. Water Content of BC-GLE hydrogel composite measurement

A 2x2x1 cm BC-GLE hydrogel composite sample was weighed using an analytical balance to obtain wet weight ( $W_w$ ). Then, it was flattened by using glass, and fed into an oven with a temperature of  $\pm 105^\circ\text{C}$ . The BC-GLE hydrogel composite was weighed every 15 minutes to obtain a constant final weight ( $W_d$ ). The same method was used to calculate the water content of pure BC. The percentage of water content can be calculated using the formula:

$$W_c(\%) = \frac{W_w - W_d}{W_w} \times 100$$

where,  $W_c$  = water content (%),  $W_w$  = wet weight (g), and  $W_d$  = dry weight (g).

The water content of GLE was also measured by putting 10 mL of GLE into hot vapor plate and weighed to obtain the initial weight ( $W_i$ ). Then, the vapor plate containing GLE was fed into the oven and weighed every 1 hour until a constant weight ( $W_k$ ) was obtained. The percentage of GLE water content can be calculated using the formula:

$$W_c(\%) = \frac{W_i - W_k}{W_i} \times 100$$

where,  $W_c$  = water content (%),  $W_i$  = initial weight (g) and  $W_k$  = dry weight (g).

##### 2.6.2. Compressive strength of BC-GLE hydrogel composite measurement

A 2x2x1 cm BC-GLE hydrogel composite sample was placed on a 4x4x4 cm concrete block that serves as a supporting material. BC-GLE hydrogel composite sample and concrete block was placed inside Tony Technik Compressive Tester's test chamber, and then the equipment was operated. The BC-GLE hydrogel composite sample was observed until the sample was flattened. The compressive force required to flake the sample would be visible when the pressure device began to press the surface of the concrete block resulting in a dramatic increase in compressive forces and in the event of an increase in this process the suppression is stopped. The magnitude of the compression force seen before the increment was calculated as the value of the compressive force of the BC-GLE hydrogel

composite sample. To obtain the value of compressive strength of BC-GLE hydrogel composite sample used formula:

$$P = \frac{F}{A}$$

where,  $P$  = Compressive strength ( $Pa$ ),  $F$  = Pressure ( $N$ ), and  $A$  = Area ( $m^2$ ).

### 2.6.3. Tensile strength of BC-GLE hydrogel composite measurement

The 15x2x1 BC-GLE hydrogel composite sample was flattened using glass. The sample was then pressed using an iron until it reaches a thickness of  $\pm 0.3$  mm. Next, the flattened sample was cut to the size of 15x1.5 cm, and put it on the sample holder of horizontal tensile tester. Then, the sample was pulled up, and the value of tensile and strain was obtained.

### 2.6.4. Functional group of BC-GLE hydrogel composite analysis

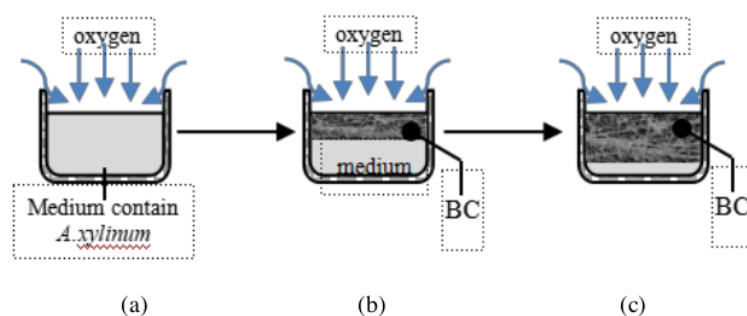
The functional group of BC-GLE hydrogel composite was analyzed by using FTIR. BC-GLE hydrogel composite sample was dried prior to analysis.

### 2.6.5. Crystallinity of BC-GLE hydrogel composite analysis

Crystallinity of BC-GLE hydrogel composite was analysed by using XRD. A dry sample of BC-GLE hydrogel composite was cut to 0.5x0.5 cm in size to obtain a flat sample portion. Sample was placed in the sample holder and placed in the XRD equipment. The obtained diffractogram was analyzed to obtain the degree of crystalline.

## 3. Results and Discussions

In the process of BC preparation, *A.xylinum* produced cellulose fibers starting from the surface to the bottom of the media solution which over time will overlap until the bacteria grow out. Illustration of BC formation can be seen in figure 1:



**Figure1.** Illustration of the formation of BC, a) initial state of the fermentation process, b) BC begins to form from the surface of the medium, c) the formation of BC continues to the bottom until the medium is depleted.

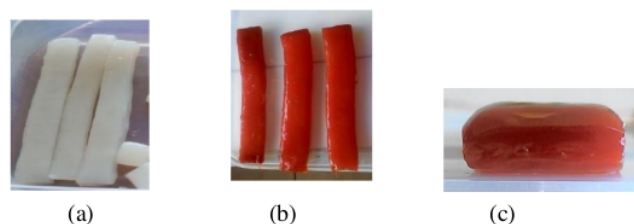
Washing and Purifying of BC was done to remove non-cellulose components and residual bacteria which was still present in BC. The remaining non-cellulose components in BC could block the hydrogen bonds that occur between the cellulose molecular chains and caused decreasing the



mechanical strength of the cellulose. The remaining bacteria also decreased the mechanical strength because these bacteria could still move on by consuming the nutrients present in the BC. Therefore, the resulting BC became fragile and could not be used to produce hydrogel composite materials that have good physical and mechanical properties [8].

The old gambir leaves used were from the Cubadak variety. One of the main compounds found in gambir leaves was Catechin. Catechin which was warmed up long enough will become catechu tannat acid due to its own condensation process. So, it becomes easily soluble in hot water [4]. Catechu tannat acid was a brownish compound commonly used as a natural dye [9]. This was what caused the resulting GLE colour brown.

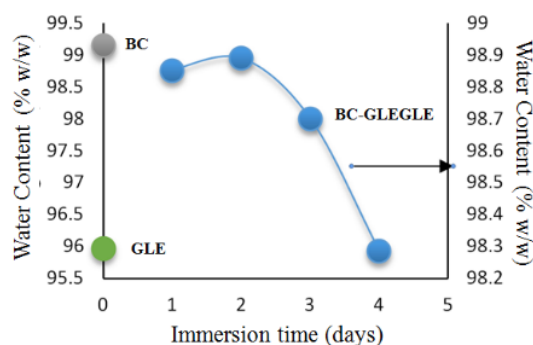
The result of immersion accompanied by stirring caused colour changed in BC due to the absorption of catechu tannat acid derived from GLE. The changes can be seen in figure 2.



**Figure 2.** Figure of pure BC hydrogel and BC-GLE hydrogel composite: a) BC hydrogel before immersion to GLE, b) BC-GLE hydrogel composite formed after immersion to GLE, c) Side view of BC-GLE hydrogel composite

### 3.1 Water Content of BC-GLE hydrogel composite

The water content is the total amount of water contained in a material compared to the dry weight of the material. The water content was a physical properties test parameter to determine how much water was contained within the BC-GLE hydrogel composite during the immersion time and how much GLE was absorbed to replace water in BC. Water content measurements were also performed on GLE to determine the percentage of extracts obtained during the extraction process. The effect of immersion time on water content of BC-GLE hydrogel composite can be seen in figure 3.

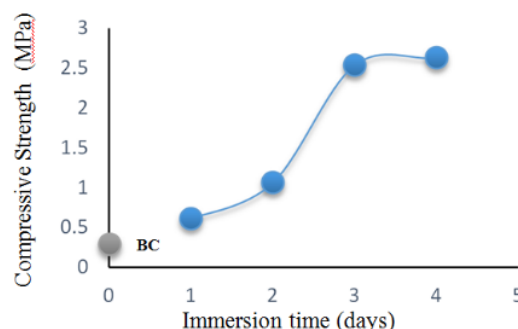


**Figure 3.** The effect of immersion time on water content of BC-GLE hydrogel composite

BC is a material that has a high water content exceeding 90% of its total weight [10]. From the figure 3, the water content of BC was 99.156%, while the water content in GLE was 95.970%. The water content in the BC-GLE hydrogel composite tends to decrease with the length of the immersion time. Water content reduction was caused by the absorption process of GLE which was acting as absorption filler replaces the water within the BC (matrix).

### 3.2. Compressive strength of BC-GLE hydrogel composite

Compressive strength is one of the test parameters of mechanical properties performed to determine the durability of a material when given the compressive force. It is necessary to know the compressive strength of the resulting BC-GLE hydrogel composite to be comparable to the compressive strength of the cartilage. The effect of immersion time of BC-GLE hydrogel composite on the compressive strength value can be seen in figure 4.



**Figure 4.** The effect of immersion time on the compressive strength of BC-GLE hydrogel composite

Figure 4 showed an increasing in compressive strength value along with the BC-GLE hydrogel composite immersion time. The best compressive strength value reached 9 times higher than that of pure BC which was obtained on the 4<sup>th</sup> day of BC-GLE hydrogel composite (2,633 MPa). This is mean that the incorporation of BC with other materials into composite material proved to increase the compressive strength of BC [1].

The increasing value of BC-GLE hydrogel composite compressive strength was caused by the amount of filler absorbed into the BC matrix. Filler replaced the position of water that filled the cavity on the BC network. It could be said that the longer the immersion time, the more filler was absorbed, so that greater compressive force was needed to flake the BC-GLE hydrogel composite. The occurrence of absorption or absorption process was also evidenced by testing the water content. The compressive strength value possessed by the BC-GLE hydrogel composite was still below the cartilage compressive strength value of 14-59 MPa [11].

### 3.3. Tensile strength of BC-GLE hydrogel composite

Tensile strength testing is the maximum tensile force a material can retain during the test until the material is disconnected. Tensile strength testing is useful to determine the elasticity of BC-GLE hydrogel composite which can be determined through the comparison of the value of tensile strength (stress) with strain value. The elasticity value is directly proportional to the tensile strength value of a

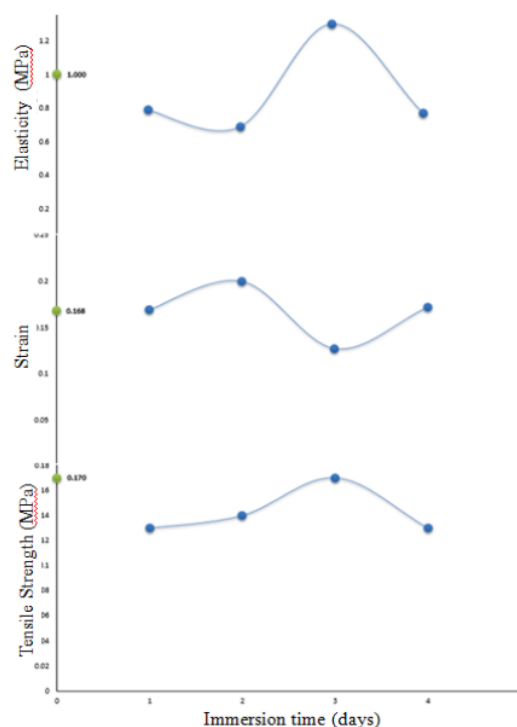


material and is inversely proportional to the strain value. The graph of time immersion ratio to the value of tensile strength, strain and elasticity can be seen in figure 5.

Figure 5 showed that the value of BC's tensile strength was 0.170 MPa. The value of tensile strength of BC-GLE hydrogel composite increased with increasing immersion time. The highest value of tensile strength was found in the 3<sup>rd</sup> day of immersion time. However, the value of tensile strength decreased during submersion entering the 4<sup>th</sup> day. This phenomena happen was due to the amount of GLE as a filler contained in the BC on the 1<sup>st</sup> and 2<sup>nd</sup> days was still low, so it did not significantly affect the resistance of the sample when it was withdrawn. Meanwhile, on the 3<sup>rd</sup> day, the amount of GLE absorbed substantially replaced the water in the BC matrix cavities, causing increased of tensile strength value in the BC-GLE hydrogel composite.

Because the interaction between filler and matrix was not accompanied by a bond formation between the two, but only the process of physical absorption, then on the 4<sup>th</sup> day, filler contained in the matrix regardless. It was also affected by the shock generated by the shaker that helps the release of the filler from the matrix so that the sample tensile strength drops again. Drop in tensile strength value was evidenced by the significant reduction of water content from the 3<sup>rd</sup> day BC-GLE hydrogel composite to the 4<sup>th</sup> day which means that the amount of filler in BC was decreased considerably.

A long with the value of tensile strength, the elasticity of BC-GLE hydrogel composite on the 1<sup>st</sup> and 2<sup>nd</sup> day tends to decrease, but increased on the 3<sup>rd</sup> day (1.3 M.Pa) and again decreased back on the 4<sup>th</sup> day. This was related to the amount of filler present in the sample matrix.



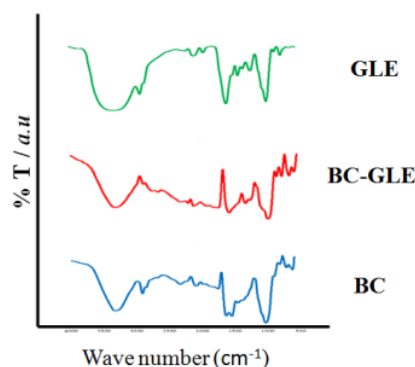
**Figure 5.** The effect of immersion time on the tensile strength of BC-GLE hydrogel composite

### 3.4 Functional group of BC-GLE

The BC-GLE hydrogel composite functional group test was performed using FTIR instrument. The spectra generated from the test were analyzed qualitatively to see if a new functional group was formed after merging BC with GLE. It aimed to know the interaction that occurs between the two during the immersion process that involved changing in physical and mechanical properties characteristics of BC-GLE hydrogel composite. IR spectra of BC, GLE and BC-GLE hydrogel composite can be seen in figure 6.

The peak cellulose characteristics were shown in the wave number range of  $4000\text{--}600\text{ cm}^{-1}$  as shown in Figure. 6. The characteristic peak of the functional group of BC was in the strain vibration OH ( $3100\text{--}3800\text{ cm}^{-1}$ ), CH ( $2901\text{ cm}^{-1}$ ), CO ( $1370\text{ cm}^{-1}$ ) and COC ( $1163\text{ cm}^{-1}$  and  $1068\text{ cm}^{-1}$ ) [12] [13].

Figure 6 showed that in BC, there was a wide peak at a wavelength of  $3335.85\text{ cm}^{-1}$  indicating a vibration of a group of OH, a vibration at a wavelength of  $1635.11\text{ cm}^{-1}$  indicating the existence of a six cycle ring of glucose monomers,  $1033.81\text{ cm}^{-1}$  indicating the presence of a  $\beta$ -1,4-glycosidic COC bond. This was the characteristic peaks of cellulose [14].



**Figure 6.** FTIR spectra of BC, GLE and BC-GLE hydrogel composite

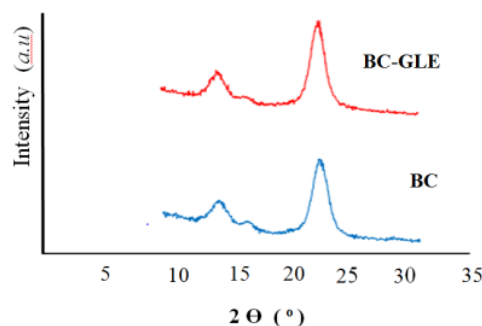
Based on the data obtained, the interaction between GLE and BC did not produce a new peak, but only caused shifting in the functional group vibration to a lower wave number. This shift proved that GLE appeared in BC-GLE hydrogel composite. So, it could be said that the interaction that occurs between GLE and BC was only physical interaction that was not accompanied by the formation of bond between the two.

### 3.5 Degree of crystallinity of BC-GLE hydrogel composite

The degree of crystalline was performed using XRD. The Diffractogram of BC-GLE hydrogel composite showed that the type cellulose produced was cellulose type I which was present at  $2\theta$   $14^\circ$ ,  $16^\circ$ ,  $22^\circ$  and  $22.48^\circ$  [15]. Diffractogram of BC and BC-GLE hydrogel composite can be seen in figure 7.

Based on figure 7, there was no significant difference occurred from the BC and BC-GLE hydrogel composite diffractogram. The absence of new emerging peaks indicated the absence of bonding between BC and GLE, but only the absorption of physics. To determine the degree of crystalline, the diffractogram was analyzed manually by transferring the diffractogram onto the graph paper and measuring the peak height as well as weighing the crystal and amorphous portions to obtain the weight

ratio of crystals and total weight (amorphous + crystals) [16]. The result of calculation of crystalline degree of BC-GLE hydrogel composite can be seen in table 1.



**Figure 7.** Diffractogram of BC and BC-GLE hydrogel composite

The results obtained that the BC-GLE hydrogel composite had a degree of crystalline greater than BC with an amorphous structure of only 32.414%. The measurement of the degree of crystalline showed that BC had a larger amorphous structure, so that BC was slightly more elastic compared to BC-GLE hydrogel composite.

**Table1.** Degree of Crystalline of BC-GLE hydrogel composite

Samples	Total weight (g)	Crystalline (g)	Amorphous (g)	Degree of crystalline (%)
BC	0.222	0.149	0.073	67.117
BC-GLE	0.203	0.137	0.066	67.586

#### 4. Conclusion

The composite hydrogel based on BC-GLE can be synthesized. The Water content of BC-GLE hydrogel composite was found lower than pure BC. Immersion time of BC in the GLE to produce BC-GLE hydrogel composite improved the physical, mechanical (compressive and tensile strength) properties of the BC-GLE composite hydrogel compare to pure BC. However, these improvements were not suitable for replacement of cartilage yet.

#### Acknowledgment

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