Effect of Adding of Green Betel Leaf Extract and Honey on Mechanical Properties of Chitosan/PVA Hydrogel Wound Plaster

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Abstract. Wound plasters on the market still have drawbacks, including lack of elasticity and stinging. Thus, there is a need for new materials from nature to be used as the base material for wound plasters, namely betel leaf extract and honey composited with PVA/chitosan. This study purpose to find out mechanical properties of wound plaster of chitosan/PVA with the composition of betel leaf extract and honey. The first stage is making hydrogel by mixing PVA and chitosan, and the second stage is extracting green betel leaves and honey by the maceration method. Then the two stages were mixed with a composition of 5wt%, 15wt%, and 25wt% green betel leaf extract. The results were tested for tensile strength to determine the elasticity properties of the wound plaster. In this study, the 25wt% betel leaf extract concentration had the best tensile strength value of 0.219 MPa, and the modulus of elasticity was 0.11 MPa.

1. Introduction
As a human organ in a protective layer, the skin is very susceptible to injury, such as wounds from stab wounds, burns, surgery or other causes. The effect can cause pain and scarring around the wound. Wound treatment is done by covered, one of which is using wound plaster. Through plaster, wound care will be more practical, comfortable and minimize contamination with the surrounding environment. However, wound plasters that are widely sold in the market still have drawbacks such as soreness and lack of elasticity so that when they are replaced with new plasters, they still cause wounds around the area covered by plaster, so there is a need for a better model of wound plaster such as hydrogel wound plaster. Hydrogel is a polymer wound dressing that is transparent, flexible, soft, and non-irritating [1] due to the release of compounds, namely as a transport medium. Currently, hydrogel wound plasters based on PVA, chitosan and starch have been developed [2]. Polyvinyl alcohol and chitosan are synthetic polymers as the main ingredients of hydrogels.

Chitosan has a high solubility in weak acids and high hydrophilicity based on amino and hydroxyl groups in the chitosan framework [3]. This chitosan can come in different forms and structures of materials such as hydrogel [4], film [5], nanofiber [6], and a 3D porous scaffold [7]. However, this hydrophilicity has an impact on low mechanical properties. So there needs to be modifications to reduce
the shortcomings, such as using blending, composite and grafting techniques [8]. It is consistent with previous research that chitosan hydrogels have toughness and are shrinkable fast [9].

Consequently, the utilization of chitosan hydrogel in biomedical materials is still very limited [10]. In addition, other research has also indicated that the mechanical characteristic of the chitosan biopolymer after the chemical modification process can be significantly improved and combined with other polymers that are more effective for wound healing as wound dressings or plasters [11]. Polyvinyl alcohol (PVA) is one of the polymers used as a blending material for chitosan to improve thermal and mechanical stability. PVA properties, such as being easy to dissolve in water, mechanical stability and flexibility, easy to form into films and non-toxic, are the basis for using PVA for applications in the medical field [12].

Chitosan/PVA blending has been extensively studied and developed in various applications. The characteristics of the chitosan/PVA blend material that have been carried out include mechanical, thermal, morphological and pH sensitivity [13]. Crosslinkers are used in the manufacture of chitosan/PVA blends to improve their mechanical properties. The crosslinking material is glutaraldehyde (GD). Glutaraldehyde functions as a filler in the chitosan/PVA matrix. This material is usually applied to drug delivery systems. The results obtained represent that the amount of filler upgrades the mechanical stability of the matrix [14].

Besides GD, other materials used as fillers are essential oils and oxalic acid [15]. Many essential oils are contained in betel leaves, while oxalic acid can be found in honey. Previous studies have shown that essential oil has been shown to affect the physical properties of the wound plaster edible film [15]. Betel leaf and honey and improving the mechanical properties of chitosan/PVA blending can also minimize infection or other effects on wounds caused by synthetic materials because they are made more natural. This research aims to escalate the mechanical characteristic of the chitosan/PVA composite material with green betel leaf extract as filler and honey. The filler composition used is 5wt%, 15wt%, and 25wt%. It is hoped that the wound plaster can increase its elastic properties to minimize the emergence of new wounds when changing the plaster.

2. Method

2.1. Materials
The materials used in this research included green betel leaf, PVA, chitosan, honey, acetic acid (Merck), water demineralized, ethanol 96% (Merck), and aluminium foil.

2.2. Preparation of chitosan/PVA hydrogel wound plaster with green betel leaf extract and honey

Green betel leaves were washed with distilled water and then dried, and the texture of dry leaves was obtained. Dried green betel leaves were crushed using a blender, and the result was a fine powder sifted using a 200 mesh sieve. Then weighed and soaked with ethanol solution where the concentration of green betel leaf simplicia in ethanol solution was 5wt%, 15wt% and 25wt% respectively and soaked for three days. After the maceration process, the product was filtered and evaporated at 80°C for 4 hours. In addition to green betel leaf, honey is also extracted with a concentration of 100 % in the same way as betel leaf extraction.

Three grams of PVA were mixed in 100 ml of water demineralized and then stirring at 120°C. One percent acetic acid/demineralized water solution was mixed with 3 grams of chitosan and then homogenized at 70°C. The PVA and chitosan solutions were combined with the composition ratio of PVA: chitosan (3:1) stirred until the viscosity increased and green betel leaf extract and honey were added with the ratio of honey and betel leaf composition (1:1) and stirred again until homogeneous. The homogeneous membrane material was placed into a petri dish and oven at 85°C during 5 hours and cooled at room temperature.

2.3. Research Flowchart
The research stages used are outlined in the flowchart as Figure 1.
2.4. Instrumentation

Analysis of elasticity properties of chitosan/PVA hydrogel wound plaster with the addition of green betel leaf extract and honey using a tensile strength test at a speed configuration of 10 mm/minute using the SHIMADZU AG-10-TE10 TE autograph. The sample to be tested was cut to a size of 1x6 cm. Characterization of tensile strength gives results in the form of a graph of stress versus strain which is then analyzed and provides information in the tensile strength (MPa), modulus of elasticity (MPa). The sample tested was in the form of edible film hydrogel, where betel leaf extract and honey were homogenized with chitosan/PVA then evaporated at a specific temperature. The sample as tested by applying a special force (pull) until the sample stretches until the sample is disconnected. Analysis for tensile strength use equation (1) and elastic modulus value use in equation (2) [16].

\[ F = \frac{P}{A} \]  
\[ E = \frac{\sigma}{\varepsilon} \]

- \( F \): Tensile strength (MPa)
- \( P \): Tensile force (N)
- \( A \): Cross-sectional area (mm²)
- \( E \): Modulus of elasticity (MPa)
- \( \sigma \): Tensile strength (MPa)
- \( \varepsilon \): Strain

Surface structure analysis with green betel leaf extract and honey with the SEM test. The SEM test procedure, the best sample from the tensile strength test results, is placed on a round copper metal plate (sample holder). Furthermore, a coating process is carried out with a gold layer so that the sample has conductive properties. The SEM (Scanning Electron Microscope) test was carried out using the HITACHI FLEXSEM 1000 tool. The sample to be tested was cut to a size of 1x1 cm.

![Figure 1. Flowchart of research stages](image-url)
3. Results and Discussion

The physical appearance of the chitosan/PVA hydrogel with green betel leaf extract and honey looks transparent and greenish, where the higher the concentration of green betel leaf extract, the greener the colour is, as shown in Figure 2. The higher the concentration of betel leaf extract, the more betel leaf content, such as essential oil, where during the process of making green betel leaf extract, the amount used also increases according to the concentration. Membrane printing was done by evaporating the solvent at a temperature of 85°C, the transition temperature of PVA glass [16]. At this temperature, PVA will form bond hydrogen between the hydroxyl group of PVA and the carbonyl group of the amide resulting from a blend of chitosan and acetic acid. In addition, PVA, which functions as an interpenetrating network (IPN), will fill the hydrogel cavity so that the structure becomes tight and causes its mechanical strength to be more assertive when compared to pure chitosan. Based on the results of tensile strength measurements on chitosan/PVA wound plaster with betel leaf extract and honey with different concentrations of betel leaf extract, which can be observed in Table 1.

![Figure 2. Chitosan/PVA hydrogel with green betel leaf extract and honey](image)

The tensile strength test results in Table 1 show the best results in the sample with a concentration of green betel leaf extract and honey of 25wt% with a tensile strength value of 0.219 MPa and an elastic modulus of 0.111 MPa. It indicates that hydrogel wound plaster with betel leaf extract and honey on a concentration of 25wt% green betel leaf extract is quite elastic and not easily torn. In addition, compared with several other hydrogels, wound plasters such as carrageenan PVP hydrogel wound plaster with a tensile strength value of 0.193 MPa [18] and polydopamine/polyacrylamide hydrogel wound plaster with a tensile strength value of 0.051 MPa [19]. So that the chitosan/PVA hydrogel wound plaster with betel leaf extract and honey is a good composition for elastic wound plasters, safe and comfortable when applied and does not cause new wounds when changing new plasters.

| Sample concentration | $\Delta L$ (mm$^2$) | Force (N) | Tensile strength (MPa) | Strain | Modulus of elasticity (MPa) |
|----------------------|---------------------|-----------|------------------------|--------|-----------------------------|
| 5 wt%                | 39.066              | 0.256     | 0.102                  | 0.928  | 0.110                       |
| 15 wt%               | 58.672              | 0.460     | 0.138                  | 1.314  | 0.105                       |
| 25 wt%               | 80.949              | 0.526     | 0.219                  | 1.973  | 0.111                       |

The mechanical properties of a material can be analysed using a tensile test, wherewith this test, the elasticity value of edible plaster in hydrogel wound cab be determined. A high modulus of elasticity and
tensile strength can protect the product from mechanical disturbances and is more comfortable to use. When the force applied to the hydrogel wound plaster of betel leaf extract and honey is large while the value of the increase in length is also more significant, it means that the wound plaster is relatively elastic, and it is proven that the hydrogel wound plaster film with a concentration of 25wt% green betel leaf extract can withstand a force of 0.526 N with an increase in length of 80.949 mm. The greater concentration of the sample in the hydrogel in Table 1, the greater the tensile strength value and the greater the elasticity. It indicated the increase in the value of delta L or the change in length in the sample being tested. This increase in length was due to the sample concentration of 25wt% having more betel leaf extract composition than the other samples, then adding betel leaf extract and honey to the chitosan/PVA hydrogel wound plaster could improve its mechanical properties. Surface morphology as well as pore formation and connectivity between hydrogel pores are very important for hydrogel wound plaster application in the formation of new tissues which can be observed through the SEM test [20].

Figure 3. SEM test results of hydrogel wound dressing, green betel leaf extract and honey with a concentration of 25 wt% enlargement (a) 500x (b) 1000x (c) 2000x (d) 3000x

Figure 3 shows the morphological structure of the hydrogel surface, which is porous, almost flat, smooth, and regular and there are few air bubbles. The presence of these pores indicates that the addition of green betel leaf extract concentration results in a larger surface area and is able to absorb excess exudate in the wound. The porous structure ensures good permeability, which is beneficial for wound healing. Meanwhile, the formation of air bubbles resulted in the mechanical properties of the hydrogel wound plaster getting smaller and more prominent.

Green betel leaf extract can permeate water and intensify the viscosity of the chitosan/PVA matrix. This viscosity affects its elastic properties, and the physical appearance of the wound plaster which is too runny cannot meet commercial requirements. The viscosity of the chitosan/PVA matrix will cause also the hydrogel wound plaster to be drier and break easily. The augment of betel leaf extract and honey increased the crystalline structure of the hydrogel [20]. Here, polymer chain arrangement occurs, filling the hydrogel's amorphous region with green betel leaf extract. When the hydrogel was given a tensile force, the first to receive the force is the amorphous region, and only then this crystalline structure will withstand the given tensile force to the maximum extent. Therefore, the more crystalline regions, the greater the hydrogel mechanical strength. The volatile oil compounds in green betel leaf extract that are soluble in water will strengthen the intermolecular bonds of the hydrogel wound plaster, causing the
tensile strength value of the hydrogel wound plaster to strengthen. Following previous studies, adding essential oils into edible films with higher concentrations will affect its mechanical properties [20].

The increase in the elasticity value of a hydrogel film, according to Iqbal et al. [21], is due to the polymer matrix used, which can increase the formation of hydrogen bonds and the strengthening effect, causing the success of the polymer itself in forming hydrogen bonds so that the content contained in the green betel leaf extract can form high hydrogen bonds in the wound plaster film and has increased the elongation value when the polymer reaches its breaking point.

4. Conclusion
The results of this research that addition of green betel leaf extract and honey to chitosan/PVA hydrogel wound plaster affects its mechanical properties, including tensile strength test. The most excellent tensile strength value was obtained in the hydrogel with the most significant green betel leaf extract concentration, namely 25% with a tensile strength value of 0.219 MPa. There is a need for further research on chitosan/PVA hydrogel wound plaster with several tests carried out such as SEM and anti-bacterial to determine the effectiveness of chitosan/PVA hydrogel with betel leaf extract and honey as wound plaster.

Acknowledgement
The researcher is grateful to the material laboratory at the Department of Physics, Universitas Negeri Surabaya, as a place for material synthesis and thanks to the Physics Laboratory of Universitas Airlangga as a place for mechanical testing of the results of this research.

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