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# Optimization of optical properties of $\text{Ba}_{0.2}\text{Sr}_{0.8}\text{TiO}_3$ thin films for a glucose sensor implementation

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**Abstract.** The development of non-invasive blood glucose measuring devices continues to be developed. The photosensor is one of the instruments for detection the blood glucose, and the high performance of sensor is depending on the optical properties. Consequently, the observation about optical properties of  $\text{Ba}_{0.2}\text{Sr}_{0.8}\text{TiO}_3$  (Barium Strontium Titanate) is important. In this paper, a solution of  $\text{Ba}_{0.2}\text{Sr}_{0.8}\text{TiO}_3$  (Barium Strontium Titanate) with chlorophyll extract from spinach and papaya leaves has been deposited on the p-type Si (100) by spin coating technique. This research aims to find the effects of presence of chlorophyll in the  $\text{Ba}_{0.2}\text{Sr}_{0.8}\text{TiO}_3$  films according to their optical properties and phonon mode. The film fabricated consisting of BST, BST with 2.5% spinach chlorophyll, BST with 5% spinach chlorophyll, BST with 2.5% papaya chlorophyll, and BST with 5% papaya chlorophyll. Therefore, the characterization was using UV-Vis spectrophotometer and micro Raman spectroscopy. The result showed that the addition of papaya chlorophyll have the optimal character than spinach chlorophyll. BST with papaya chlorophyll showed a significant decrease in the energy gap. Also, it has a phonon mode with a Raman shift that supports the result.

Keywords:  $\text{Ba}_{0.2}\text{Sr}_{0.8}\text{TiO}_3$  films, chlorophyll, spin coating, UV-Vis, Micro-Raman

## 1. Introduction

Globally 4.6 million cases of death occur each year due to diabetes. Children and adolescents died of insulin deficiency without being diagnosed [22]. Diabetes mellitus is a disease caused by increasing the blood glucose levels due to the disruption of the metabolic system, where the pancreatic organs are unable to produce the insulin hormone according to the body's needs [19]. If a wound occurs, then the blood is difficult to stop so the wound becomes more severe and rot. Measurement of blood glucose levels by taking a blood sample is very risky if the scar does not dry out immediately. For this reason, the development of non-invasive blood glucose measuring devices continues to be developed. Non-invasive measurements on this device utilize optical phenomena in the form of light absorption at specific

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Fig. 1. Blood glucose measuring tools [3].

wavelengths of blood glucose (visible light 534 nm and infrared 939 to 2326 nm). The amount of this absorption depends on the concentration of glucose in the bloodstream [3].

In some countries, biosensor as a measurement of blood glucose continues to be developed. The photo-sensor is one of the tools used for detection of blood glucose levels. So, we need the instrument for measuring the intensity of the light captured after passing through the tip of the finger. Because one of the organs of the human body that is easily penetrated by light is the fingertips (Fig. 1) [2].

One of the materials that can be used as a sensor is Barium Strontium Titanate (BST) [15]. Irzaman et al., (2016) explained that the BST ferroelectric material can be used for light sensor applications and can be developed into solar cells. BST is a semiconductor material that has been researched in recent years for the various application, especially many types of sensors [8,12,17,18,24,38].

The performance of barium strontium titanate ( $Ba_{1-x}Sr_xTiO_3$ ) thin film is excellent in applications of tunable microwave devices, fast nonlinear optics, and high-density gigabit dynamic random access memory [34]. The physical properties of BST films are strongly influenced by composition, dopants, deposition, internal stresses, electronic structures, and others. Currently, the development of BST sensor is continuously carried out to increase sensitivity or accuracy in the measurement process [8].

In Indonesia, the application of chlorophyll as an energy source is being pursued because chlorophyll is found in all photosynthetic plant so chlorophyll is easy to find out [6,36]. All of chlorophyll can fluorescence, if it gets irradiated with a particular spectrum of light (excitation spectrum), then the light it passes on (emission spectrum) is light on a different spectrum. For example, chlorophyll-a dissolved in acetone 85% has a maximum excitation between wavelengths of 430–450 nm (blue-purple) and will give a maximum emission between wavelengths of 650–675 nm (dark red) [40]. In material science, it is usually applied as an energy storage and detector material. For the example [18] use chlorophyll as Thiourea doping for the active potential of the photovoltaic cell layer [1].

In this paper, we compare the chlorophyll extracts taken from several plants. The leaves investigations were spinach and papaya leaves. In previous research, chlorophyll extract is known to have the ability to absorb light. By using UV-Vis and Micro-Raman Spectroscopy, we analyzed the differences in BST thin film with spinach and papaya chlorophyll extract. BST films without doping were also presented as a control sample.

### 1.1. Chlorophyll

Pigments or dyes, generally found in meristem tissue cells which form a chloroplast. Chloroplast has very diverse shapes and sizes [39]. Chloroplast is composed of a stroma, covered by membranes,

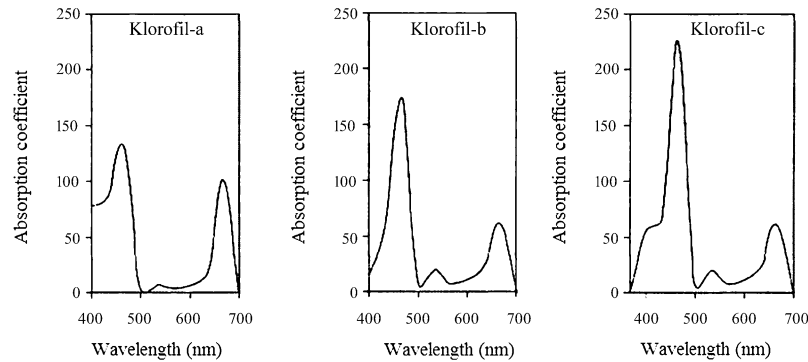


Fig. 2. The absorption spectrum of chlorophyll-a, -b, and -c uses ether solvents [30].

in which small granules are scattered containing green chlorophyll pigments. Pigments in chloroplast (chlorophyll) have an essential role in the photosynthesis, it is a catalyst and absorbs the kinetic energy [33]. From the research conducted by Meyer and Anderson (1952), chlorophyll is divided into 3 types, chlorophyll-a, -b and -c. In this study, chlorophyll-a, -b and -c have the same chemical properties which are insoluble in water but can dissolve in various types of organic solvents. *Chlorophyll-a* is soluble in ethyl alcohol, ethyl ether, acetone, chloroform, and carbon-bisulfide, chlorophyll-b and -c also soluble with them but it is not as easy as chlorophyll-a [30]. Besides, if *chlorophyll-a* and *chlorophyll-b* are hydrolyzed, alcohol (phytol) will be obtained. The phytol group forms a third of the chlorophyll molecule and has a strong affinity for oxygen. If it is ignored, pure chlorophyll will leave a residue composed only of magnesium-oxide. Although iron (Fe) and other minerals are essential for the formation of chlorophyll in living cells, magnesium (Mg) is the only metal element as a catalyst in visible light absorption [30].

Based on the Yentsch and Menzel (1963) study, all of the chlorophyll have the fluorescence properties, when it get irradiated with a certain spectrum of light (excitation spectrum), the light is transmitted (emission spectrum) in a different spectrum. For example, chlorophyll-a dissolved in 85% acetone has a maximum excitation between wavelengths of 430–450 nm (blue-purple) and will provide maximum emission between wavelengths of 650–675 nm (dark red). The absorption spectrum of chlorophyll-a, -b and -c in ether solution is shown in Fig. 2, the maximum absorption of primers is in the blue-purple spectrum (wavelength 430–477 nm) and secondary maximum in the red spectrum (628–662 nm) [30].

### 1.2. UV-Vis and Raman spectroscopy

UV-Vis Spectrophotometry used to analyze the energy gap of thin film. Spectrophotometric measurements involve considerable electronic energy in the molecules analyzed, so the UV-Vis spectrophotometer is more widely used for quantitative analysis than qualitative [20]. This is appropriate with the measurement range desired by this study. Energy gap is obtained by drawing a line of the relationship between  $[\ln(\frac{R_{\max}-R_{\min}}{R-R_{\min}})]^2$  to  $h\nu$ . The value that cuts the  $h\nu$  axis is the energy gap value [10,13,14].

Another instrument is Micro-Raman Spectroscopy, which used to analyze samples (solid, liquid, even biological samples). Micro-Raman Spectroscopy has an accurate level of measurement, non-destructive to the sample, so the analysis does not damage the physical sample [23,32]. In a previous study conducted by S. Y. Wang, 2006, Raman spectroscopy was used for measuring thin films doped using Cerium. The results of his research stated that Raman Spectroscopy can analyze the crystal structure, surface morphology, and stability of the Ce-doped BST film [37]. Micro-Raman Spectroscopy type in this research

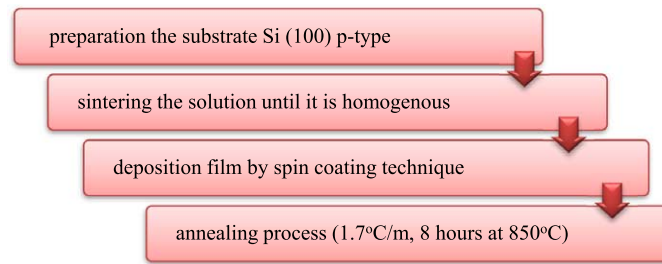


Fig. 3. Fabrication of BST and BST-Chlorophyll thin film.

using *DU420-BROD semicolon (indoortechnology co. Ltd., northern Ireland)* because this instrument is able to measure the sample at any condition, solid as solution alike [32].

## 2. Materials and experimental procedures

### 2.1. Materials

The material needed is Barium acetate [ $Ba(CH_3COO)_2$ ], Strontium acetate [ $Sr(CH_3COO)_2$ ], Titanium Isopropoxide [ $Ti(C_{12}O_4H_{28})$ ], Si (100) p-type, ethylene glycol, acetic acid, and aquadest. In this study, the ratio of solvent was 3:1 between acetic acid and ethylene glycol.

### 2.2. Experimental procedures

The first procedures is chlorophyll extraction, The extraction process is starting on cleaning the leaf with the running water, throwing away the leaf bones, then blending 2 part of leaf with 1 part of aquadest, after that filtering the solution and blanching at  $60^\circ C$  for 10 minutes, collecting the clumps, moving into the glass ware petri, and drying at  $50^\circ C$ .

The second is fabrication of BST and BST-Chlorophyll thin film. It was following the research conducted by Irzaman [13], as seen in Fig. 3.

## 3. Results and discussion

The optical properties was characterized by UV-Vis spectroscopy. The reflectance mode can be used to estimate the energy gap. Energy gap is energy needed by electron to excite from the valence band to the conduction band [9,16]. The lowest energy is mean the easiest of electron to move [11,21]. According to the research conducted by Ruzimuradov 2007, BST has 3.2 eV of energy gap [31]. In this study found that BST have 2.82 eV, it was close and less than the reference. The analysis result can be seen in Fig. 4 and Fig. 5, also summarized in Table 1.

The reflectance as an energy is shown in Fig. 4. Based on the UV-Vis characterization, BST and BST-chlorophyll film in this research have the energy gap smaller than the reference. In Fig. 5, the energy gap can be seen from the change in potential energy or the slope of the line. If the slope of the line gets bigger, the energy gap will be even greater [4]. This research is a continuation of our previous research, BST with chlorophyll papaya, spinach, and mustard as much as 5% dissolved in acetic acid and ethylene glycol in a ratio of 3:1. The smallest energy gap found at BST with spinach 5%, it was 2.79 eV [26].

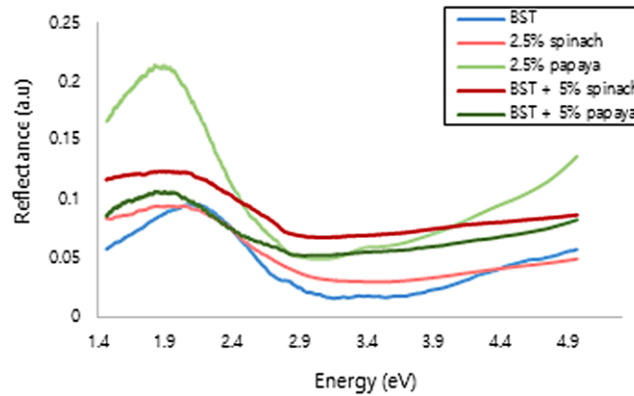
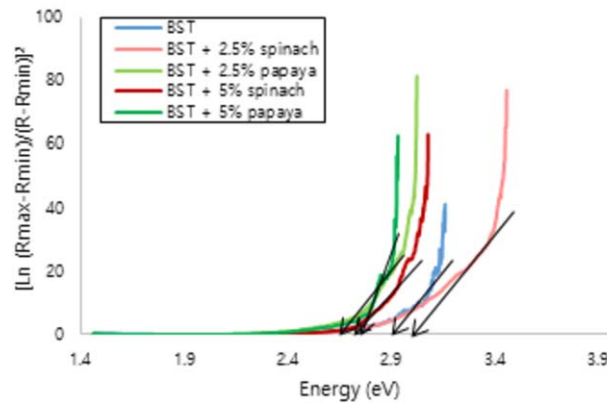
Fig. 4. Reflectance of  $Ba_{0.2}Sr_{0.8}TiO_3$  film.Fig. 5. The energy gap of  $Ba_{0.2}Sr_{0.8}TiO_3$  film.

Table 1  
The energy gap of BST and BST-Chlorophyll film

Sample	Energy Gap
BST	2.82
BST with 2.5% spinach chlorophyll	2.98
BST with 2.5% papaya chlorophyll	2.69
BST with 5% spinach chlorophyll	2.69
BST with 5% papaya chlorophyll	2.66

Also other studies on BST with 2.5% chlorophyll mustard, cassava, papaya and spinach dissolved with 3:1 have energy gaps more than 3 eV [25]. So, the composition of the solvent affects the energy gap. In this study, we use the 19:1 ratio of solvent and we got the smaller energy gaps. Besides, eventhough using the some ratio, BST with papaya in this study has smaller energy gap than BST with mustard chlorophyll [28] and BST with cassava chlorophyll in previous [27].

The Micro-Raman analysis in Fig. 6 shows that more than one phonon mode, and an increase of intensity due to chlorophyll presence starting at the wavenumber  $400\text{ cm}^{-1}$  to  $1000\text{ cm}^{-1}$ . Along with differences in chlorophyll extract spinach and papaya leaves, absorption of Transverse-Optical A(TO)/

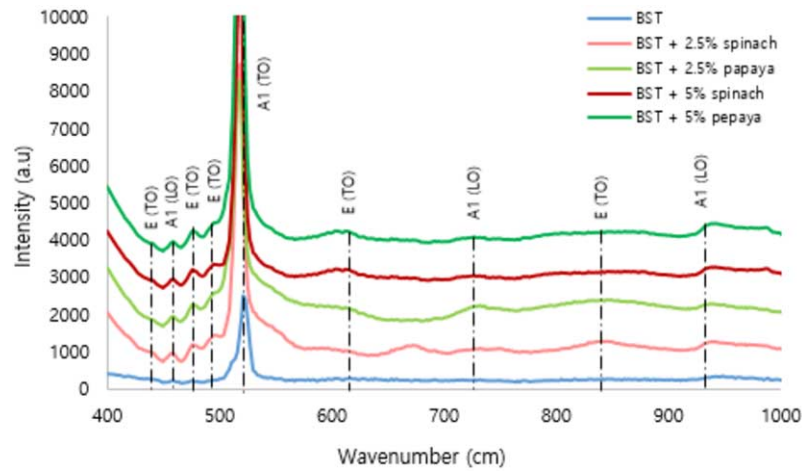


Fig. 6. Micro-Raman analysis of  $Ba_{0.2}Sr_{0.8}TiO_3$  film.

Emission of Longitudinal-Optical E(LO) Phonon [5,29] at a peak of  $523\text{ cm}^{-1}$  and E(LO) at a peak of  $676\text{ cm}^{-1}$ , it has a different shift in peak toward greater intensity. Phonon optical frequencies and symmetry modes on tetragonal BST are available in Table 2.

All of BST films with chlorophyll are perovskite structures from the tetragonal phase which is characterized by a peak intensity of  $841\text{ cm}^{-1}$  [32]. The safe intensity of BST with chlorophyll 5% papaya higher than another concentration of chlorophyll. Therefore, polycrystalline and tetragonal structures are very large. However, the low peak was shown in chlorophyll at a frequency of  $672\text{ cm}^{-1}$  is likely due to the scattering disorder activity of the TO and LO phonon branches which have a high density in the Brillouin zone boundary state [35]. The low peaks at  $947\text{ cm}^{-1}$  in all BST film samples are likely due to the residual pressure and surface pressure between the film and the silicon substrate. However, the peak at  $521\text{ cm}^{-1}$  to  $523\text{ cm}^{-1}$  in all of samples is the optical phonon mode that undergoes degeneration. At frequency of  $521\text{ cm}^{-1}$  to  $523\text{ cm}^{-1}$  the peak increase or difference in peak intensity is seen. Based on research conducted by [30], one of the factors that causes the difference in intensity at frequency  $521\text{ cm}^{-1}$  is caused by the release of the Mg atom compound, so that Chlorophyll can decompose both while still in plant cells (the operation of the chlorophyllase enzyme) or when the chlorophyll extraction process takes place [36].

Thin films which have good optical properties can be further investigated and applied as sensors. In the case of measuring blood glucose, thin film is used in devices as a capacitive biosensor. Capacitive sensor is an electronic sensor that works based on the concept of capacitive. This sensor works based on the changes in electrical energy charge that can be stored by the sensor due to changes in plate distance, changes in cross-sectional area and changes in the capacitive sensor dielectric volume. The concept of capacitors is the process of storing and releasing electrical energy in the form of electrical charges on a capacitor that is influenced by surface area, distance, and dielectric material. Capacitive sensor consists of two main components as an electrode, sensing electrode and reference electrode.

Capacitive biosensors measure the dielectric of the layer on the interface between the electrolyte and electrode. The glucose oxidase enzyme is immobilized onto electrodes to the oxidize glucose. In practice, the biochemical reaction of glucose with enzyme generates the free electron carriers. Therefore, the potential difference on the increased electrodes causing the voltage output that can be measured from the biosensor [7]. It can be concluded that this research is the beginning of a series of biosensor studies.

Table 2

Comparison of the shift mode based on the optical phonon frequency and symmetry mode on tetragonal BST

Symmetry [37]	Wavenumber (cm <sup>-1</sup> ) [37]	Wavenumber (cm <sup>-1</sup> ) (this study)				
		BST	BST + 2.5% spinach	BST + 2.5% papaya	BST + 5% spinach	BST + 5% papaya
E(TO)	416	460	459	459	459	459
A <sub>1</sub> (LO)	–	480	476	476	476	476
E(TO)	–	495	495	499	498	498
E(TO)	–	508	510	510	510	510
A <sub>1</sub> (TO)	521	523	518	518	518	518
E(TO)	676	672	673	673	674	672
A <sub>1</sub> (LO)	742	742	743	743	744	744
E(TO)	841	841	841	841	841	841
A <sub>1</sub> (LO)	947	–	947	948	947	946

Based on the analysis of optical properties and phonon modes, BST thin films have the potential to be applied as sensors, especially glucose sensors.

#### 4. Conclusion

Fabrication of Ba<sub>0.2</sub>Sr<sub>0.8</sub>TiO<sub>3</sub> film was deposited by spin coating technique. The analysis of UV-Vis spectroscopy and Micro-Raman Spectroscopy showed that the film has good characteristics. Addition of chlorophyll into BST solution raises the good interaction between chlorophyll atoms and BST. This result obtained that BST with 5% chlorophyll papaya have the smallest energy gap, and overall the addition of chlorophyll generates the small energy than the BST film without addition of chlorophyll. It can be conclude that BST thin films have the potential to be applied as sensors, especially glucose sensors.

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#### Conflict of interest

The authors have no conflict of interest to report.

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