Silica-nanoparticles in slow release supplement: preparation and characterization

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Silica-nanoparticles in slow release supplement: preparation and characterization

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Abstract. The nanosilica was prepared from bamboo leaves ash by calcination method in 800°C for an hour. The result was 350 g silica from 500 g of its ash. The SEM shows a homogeneous surface of SiO2 and the EDX consists of 56.73% Si. The FTIR test indicates the Si-O-Si functional groups. The XRD shows 2θ = 21.6 which corresponds to JCPDS No. 39-1425 and 18-1170. The supplement was vitamin C tablet which prepared into slow release supplement model, by mixing of three component: generic ascorbic acid, magnesium stearate, and nanosilica in certain composition. Some assessments were carried out to determine the tablet’s characteristics: the size, homogeneity, and hardness. Physical testing of the slow-release vitamin C tablet was found to meet the requirements of tablet by Indonesian Farmakope. In vitro assessment was represented by dissolution test in simulation solutions of stomach by 0.1M HCl and the intestine by citric acid. The result showed that the dissolution of the samples were slower than the control tablets. This indicates that the addition of silica-nanoparticles affect the characteristics of slow release supplement.

1. Introduction
Silica was found in several biomass, including the side product of agriculture. The silica in the form of nanoparticles form biomass were developed in recent years, ie from rice husk ash [1] and coconut shell [2]. Application of biomass based silica-nanoparticles were reported in the preparation of several industrial product, i.e. in food industry [3] and drug delivery [4]. The size of nanoparticles give an advantage in mobilization as nanocarrier. It was conducted in the delivery of curcumin [5]. Kumar et al, (2016) developed the silica-nanoparticles from bamboo leaf. Bamboo leaves contain more than 70% of silica [6]. This research on the preparation of oral supplement was supported by silica-nanoparticles (SNs). This paper reported the result of preliminary experiment with vitamin C as the supplement. The utilization of SNs is the part of the medical application of silica-nanoparticles as drug carrier. Slow release supplement is a model of SNs supported drug.

Vitamin C, in this case, was selected as the sample of slow release supplement. People use vitamin C as the supplement which is usually needed in increasing the body resistance. The development of slow release drug / supplement is needed to decrease the frequency of the drug/supplement consumption. The addition of supporting material as drug carrier will accommodate this purpose. One of the recent research of drug carrier in nano medicine is silica-nanoparticles (SNs) [7]. In vivo study is needed to find the bioavailability and biocompatibility in the human body, which is begin with the application in animal model [8-9].
2. Material and Method

2.1. Materials
The bamboo leaves were collected from Sleman Yogyakarta, Indonesia. The main chemicals (HCl, Sodium citrate, Magnesium Stearate, and buffer) were from Merck, vitamin C from IPI, aquadest from The Chemistry Laboratory, UNY.

2.2. Preparation of silica from bamboo ash
The bamboo ash was prepared by direct drying the leaves under the sun, followed by combustion in traditional stove, and calcined by muffle furnace at 800 °C in 6 hours. The product was then washed in 0,1 M HCl (by ratio 1:6) and well stirred in 60 minutes, and filtered by Whatman filter paper after 24 hours and then washed in water until neutral. The residue was then heated in the oven at 110 °C in 2 hours to remove the retained water [6].

2.3. Characterization of nanosilica
The morphology of the silica was determined by Scanning Electron Microscopy-Electron Dispersive X-Ray Analyser (SEM -EDX) of JEOL JED-2300 model, and then continued to determine the crystallinity by X Ray diffraction (XRD Lab-X Type 6000 Shimadzu Japan). The infrared spectrum was recorded by Fourier Transform InfraRed (FTIR) Horizon MB3000.

2.4. Preparation of BNs-supported supplement
Preparation of SNs supported supplement was conducted by mixing the materials: vitamin C, magnesium stearate and SNs in certain composition and pressed to round shaped tablets. The control sample was the mixture without SNs. The characteristic of tablets include the homogeneity, size, and hardness of the tablets. The main characteristic is the dissolution process which was determined by dissolution test in variation of media vs. time of dissolution. Dissolutions test was conducted in both HCl and sodium stearate media, as the simulation media of stomach and intestine.

3. Result and Discussion

3.1 Preparation of nanosilica from bamboo leaves
The yield from 700 grams bamboo leaves was 502 grams charcoal which was then calcined at 800°C in 6 hours to get 350 grams of greyish ash of silica.

![Figure 1. The SEM image of silica [(A= 5000x, B = 10.000x ]](image-url)

3.2 The Sem-EDX result of the nanosilica
The morphology of the silica was characterized by Scanning Electron Microscope -Electron Dispersive X-Ray analyser. The SEM image of the silica from bamboo leaves was shown in figure1. Figure 1 shows the soft and homogenous surface of silica. The particles have ellipsoid and sphere shape. Stanley et al., (2014) also reported the plate and sphere shape with the size of the aggregate wasin range of 0.5-1.0 µm [10].
The EDX result is shown in figure 2 indicating that the composition of the ash are 56.73% Si and 43.26% oxygen. The pure SiO$_2$ contained 58.63% Si, so this result was comparable to the previous publication [11].

![Figure 2. The EDX result of bamboo leaf ash.](image)

### 3.3. The FTIR Spectra of the nanosilica

The infrared spectrum of bamboo leaf silica after calcined at 600°C was shown in figure 3 and the vibration characteristic was listed in table 1. This spectrum showed the vibration of –OH, C-H, Si-O-Si, and Si-OH, which are associated with silica.

![Figure 3. The Infrared spectrum of bamboo leaf silica.](image)

| Wavenumber (cm$^{-1}$) | Vibration                                      | References                        |
|------------------------|------------------------------------------------|-----------------------------------|
| 3749.62                | Stretching Vibration of –OH form Si-OH         | Stanley and Nesaraj, 2014 [12]    |
| 2368.59                | Stretching Vibration of C-H                     | Priyanto, 2015 [13]               |
| 1095.67                | Asymmetric stretching of Si-O-Si               | Wang et al, 2000 [14]             |
| 786.96                 | Stretching Vibration of Si-O in Si-OH           | Stanley and Nesaraj, 2014 [12];   |
|                        |                                                | Priyanto, 2015 [13]               |

Table 1. The characteristic vibrations of the silica.
3.4 The X Ray Diffraction analysis of the nanosilica

The X-Ray diffraction analysis on the silica shows the pattern on the range of 2θ of 2º to 80º. The XRD pattern shows a sharp peak on 2θ=21.6º associated to Si. According to PDF No. 39-1425 and 18-1170 from JCPDS (figure 4b), Si has characteristic peaks on 2θ=20.5 º, 21.6 º, and 23.3º. The formation of crystalline silica started on 650 ºC and completed in 800 ºC. The XRD pattern of the sample was shown in Figure 4 (a) and compared with the pattern in JPPDS (b).

![Figure 4](image)

**Figure 4.** (a) The XRD pattern of bamboo leaf silica (b) The JCPDS data (reference).

The particle size was 7.057 nm, following Scherrer formula. It matches with the requirements of nanoparticles size, in range 5-100 nm. This product was labeled as Bamboo Nanosilica BNs). This product was then applied in the preparation of BNs supported supplement.

3.5 Preparation and it’s in vitro study of slow release supplement.

Preparation of BNs supported supplement was conducted by mixing the composition as shown in table 2. The mixture was then pressed to round shaped tablets with diameter of 0.72 cm. The weight of each tablet was about 160 mg. The control sample was the mixture without BNs.

| No | Sample code | composition | Vit.C | Magnesium stearate | BNs |
|----|-------------|-------------|-------|--------------------|-----|
| 1  | A           | 140 mg      | 20 mg | -                  |     |
| 2  | B           | 100 mg      | 50 mg | 10 mg              |     |
| 3  | C           | 100 mg      | 45 mg | 15 mg              |     |
| 4  | D           | 100 mg      | 40 mg | 20 mg              |     |

Table 2. Formulation of BNs supported Vitamin C.

The physical characteristic of tablets is determined in terms of size, weight, hardness, fragility and dissolved time. The result was listed in table 3.

| Physical characteristic | A      | B      | C      | D      |
|-------------------------|--------|--------|--------|--------|
| Mass (mg)               | 170.07 | 170.10 | 170.17 | 170.28 |
| Diameter (cm)           | 0.71   | 0.72   | 0.72   | 0.72   |
| Hardness (kg/cm²)       | 0.396  | 0.674  | 0.676  | 1.188  |
| Fragility (%)           | 0.2    | 0.7    | 0.8    | 1.4    |
| Dissolved time (minute) | 8.5    | 5      | 6      | 6      |

Table 3. The physical characteristic of BNs - Vitamin C.

All the result have fulfilled the requirement of oral supplement according to the 4th Indonesian Farmakope, that the standard deviation of the value is not more than 10%. The BNs addition was also found to increase the hardness of the tablets. The higher the hardness, was found the slower the supplement released. According The 4th Indonesian Farmakope, the maximum hardness of the...
supplement tablets is 1.5 kg/cm². It means, all the samples meet the requirement of the hardness. The fragility of BNs -vitamin C also meet the requirement of the standard supplement tablet, namely maximum 1%. All samples ranged in 0.4 – 0.8 %, so that the parameter of the formulation of a good tablet also has been fulfilled. The standard of dissolved time of this supplement tablet is 15 minutes (max). All samples also meet the requirements of Indonesian Farmakope [14].

The main characteristic is the dissolution process which is determined by dissolution test by variation of media vs time of dissolution. This test was associated with the ability of the drug in the interaction with the human body [15]. dissolution test was conducted in both HCl and sodium stearate media, as the simulation media of stomach and intestine. The result of the dissolution test of the sample was shown in figure 5.

Interpretation of the dissolution data was carried out by observation of the curve pattern of the formula in two media. The HCl medium is the simulation of stomach and the sodium citrate is the simulation medium of intestine. The profile of dissolution was formed by plotting of the amount of the released particles of the supplement vs time (in minute) (figure 5). The observation was carried out in the sample of 10% weight of the tablets. The rate of the release of the supplement was proportional to the BNs contained in the tablets. The higher the silica content, results in the slower the release. The standard of dissolution time of slow release drug by Indonesian National Agency of Drug and Food Control is 60 minutes. It was provided that the addition of silica-nanoparticles was useful in preparation of slow release supplement.

4. Conclusion
The preparation of bamboo leaf silica-nanoparticles (BNs) was successfully conducted by calcination of bamboo leaves ash at 800 °C in 6 hours. The particles were crystalline and the size is 7.057 in average. The utilization of bamboo leaf silica-nanoparticles in supplement preparation meet to the requirement of the slow release drug / supplement, referring to Indonesian National Agency of Drug and Food Control.

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