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Synthesis of Polyvinylpyrrolidone (PVP)-Green Tea Extract Composite Nanostructures using Electrohydrodynamic Spraying Technique

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Abstract. Green Tea Extract (GTE) as an active substance has successfully loaded to PVP nanostructures using electrohydrodynamic spraying technique. The precursor solution was the mixture of ethanolic polyvinylpyrrolidone (PVP) with a molecular weight of 1,300 kg/mol and ethanolic GTE solutions at a weight concentration of 4 wt.% and 2 wt.%, respectively, and it was estimated that the entanglement number was 2. The electrospraying was conducted at the voltage of 15 kV, the flow rate of 10 μ L/min., and the distance between the collector and the tip of the nozzle of 10 cm. The SEM images showed that the PVP/GTE nanostructures had a combination of agglomerated beads (less spherical particles) and nanofibers. This occurred because if the PVP concentration is low, the PVP/GTE composite has weak core structures that cause the shell to be easily agglomerated each other. The intermolecular interaction between PVP and GTE in the PVP/GTE nanostructures occurred as confirmed by the peak at 3396 cm^{-1} , which is the carboxyl group, proving that the PVP/GTE nanostructures contained water, alcohols, and phenols. The peak at 1040 cm^{-1} , which is the stretching of C-O group in amino acid, gave another proof to the intermolecular interaction.

Keywords: Polyvinylpyrrolidone, Green tea extract, Nanostructures, Intermolecular interaction

1. Introduction

Green Tea or *Camellia sinensis* L. is a tropical plant that is widely consumed for generations as beverages. It is beneficial for human since it has high antioxidant activity [1-3]. Moreover, it has various polyphenolic compounds containing active components known as catechins [1]. These polyphenols have been reported to have higher antioxidant activity than those vitamin C and E, water soluble, non-toxic [2], is good enough to prevent prostate and colorectal cancer [3,4], and can also extend the shelf-life of various food products [1]. However, the amount of catechins that can be absorbed by the body is just less than 1-2% when consumed orally [5]. Since the molecular weight of catechins is very high, it results in a low bioavailability [1-5]. An alternative to overcome this problem is by reducing the size of the carrier system. Several methods can be used to improve the

bioavailability of catechins or EGCC (epigallocatechin-3-gallate), which is the most abundant green tea constituent, such as encapsulation using oil-in-water sub-micrometer-sized emulsions, liposomes, and microparticles of protein/polyphenols [4]. However, for all these methods, the size is still in the micrometer scale, which must still be reduced down to the nanoscale. One way to obtain particle with nanoscale is through electrohydrodynamic spraying method.

Electrohydrodynamic spray, or famously shortened as electrospray, is an easy and expensive way to produce nanoparticles. In this technique, a high voltage is being applied to induce charges on the surface of a polymer solution. The charged polymer solution will then be attracted towards the collector by the electrostatic force to form particles or fibers [6-8]. The polymer can be used as the carrier of an active content such as green tea. Therefore, by mixing the extract of green tea with a polymer, composite nanostructures (nanoparticles or nanofibers) may be created, and it is expected that the bioavailability of green tea extract will increase.

Several studies have used polyvinylpyrrolidone (PVP) as the carrier medium in various fields of applications that involved the formation of nanoparticles or nanofibers [9-11]. Noting that PVP has been recognized by FDA (Food and Drug Administration, USA), and is widely used as a drug carrier to increase the drug solubility [10], it was then selected as the carrier of green tea extract (GTE) in the present study. To the limit of our knowledge, PVP/GTE composite nanostructures produced by using electrohydrodynamic spraying have not been reported. The morphology of the PVP and PVP/GTE composite particles was characterized using a scanning electron microscope (SEM). A Fourier transform infrared spectrometer was used to detect the presence of typical functional groups of PVP and PVP/GTE composite.

2. Experimental Method

Polyvinylpyrrolidone (PVP) with a molecular weight of 1,300 kg/mol was purchased from Sigma-Aldrich, Indonesia. Analytical grade ethanol purchased from Merck, Indonesia, was used as the solvent. Green tea extract (GTE) powder was produced using ethanolic maceration method at Laboratory of Natural Materials, ITB. PVP and GTE were dissolved in ethanol at a weight concentration of 4 wt.% and 2 wt.%, respectively. PVP solution was made as the precursor to produce PVP nanostructures through the electro spraying technique. Another PVP solution was mixed with the GTE solution and stirred for ± 2 hours at room temperature until the formation of a homogeneous solution. During electro spraying, each solution of PVP and PVP/GTE was placed in a 12-mL syringe and was connected to an applied voltage of 15 kV, a flow rate of 10 $\mu\text{L}/\text{min.}$, and at a distance of 10 cm between the needle and the collector. The electrospray was conducted at a constant humidity of 55% and at room temperature. The electrospray process lasted for approximately 5-8 hours to obtain nanostructured samples.

The viscosities, the surface tensions, and the conductivities of PVP and PVP/GTE solutions were measured using Ostwald Viscometer (Fisher), du Noüy Tensiometer (Fisher), and conductometer (Mettler-Toledo), respectively. The morphology of the nanostructures was observed using a scanning electron microscope (SEM) (Hitachi, SU3500) operating at an accelerating voltage of 25 kV with the magnification between 5,000 and 60,000 times. Meanwhile, to identify the existing functional groups and the intermolecular interactions of PVP and GTE, a Fourier transform infrared (FTIR) spectrometer (Bruker, Alpha) with the wavenumber in the range of 500 to 4000 cm^{-1} was used.

3. Results and Discussion

Syntheses of PVP nanoparticles and nanofibers were successfully conducted as reported in many previous works [8,12]. However, performing electrospray with a polymer of high molecular weight added by a bioactive substance such as green tea extract is a quite new. Electro sprayed PVP/GTE composite nanostructures were obtained using the entanglement analysis $(n_e)_{\text{soln}}$ [12]. Using ethanolic PVP and ethanolic GTE solutions at a weight concentration of 4 wt.% and 2 wt.%, respectively, it was estimated that the entanglement number $(n_e)_{\text{soln}} \sim 2$, where particles will form. Figure 1 depicts the

colors of PVP solution, GTE solution, and the mixture of PVP and GTE solutions. The white PVP solution mixed with the dark GTE solution resulted in the PVP/GTE solution with a light brown color. Two main parameters that influence the morphology and size on the electrospray are solution and process parameters [7]. The solution parameters include the concentration of polymer (as viscosity, surface tension, and conductivity), the characteristics of the solvent, and the types of additives. The process parameters include the voltage, the flow rate, the diameter of the needle and the distance between the needle and the collector. The viscosities, conductivities, and surface tensions of the PVP, GTE, and PVP/GTE solutions were measured, and the results are displayed in Table 1. The PVP solution is much more viscous than the GTE solution, and the viscosity of PVP/GTE solution is also higher than that of GTE solution but lower than that of PVP solution. The conductivity of GTE solution is higher than that of PVP solution, and therefore the conductivity of PVP/GTE solution is in between those of PVP and GTE solutions. The surface tensions of solutions have the same characteristics of their viscosities.

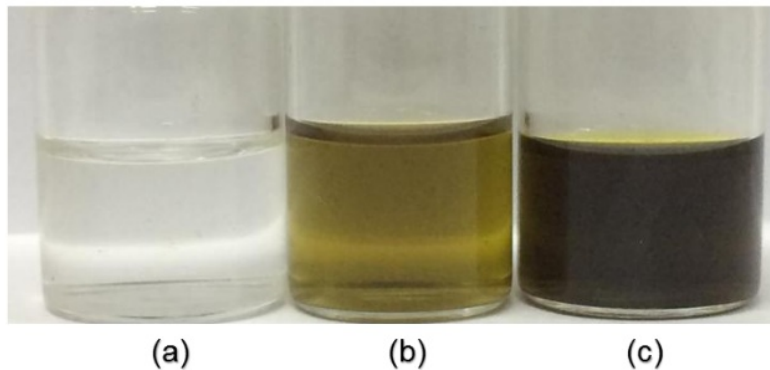


Figure 1. The colors of (a) PVP solution of 4 wt.%, (b) PVP/GTE solution with the concentration ratio of 8:2, and (c) GTE solution of 2 wt.%.

Table 1. The measurement of viscosity, conductivity, and surface tension at room temperature

Solution	Viscosity (centiPoise)	Conductivity ($\mu\text{S}/\text{cm}$)	Surface Tension (dyne/cm)
PVP 4 wt.%	7.14	0.08	29.0
GTE 2 wt.%	0.85	1.47	28.5
PVP/GTE (8:2)	3.80	0.51	28.8

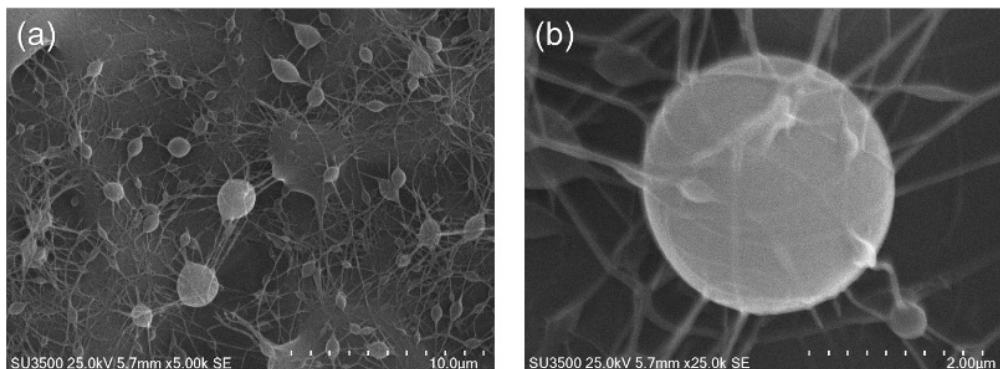


Figure 2. SEM images of PVP nanostructures being magnified at (a) 5,000 and (b) 25,000 times

The SEM images of the PVP nanostructures made by the electrospay technique are displayed in Figure 2. It was shown that the nanostructures had a combination of beads (less spherical particles) and nanofibers with a wide distribution of size. The diameter distribution of PVP beads [21] within the range of 291 to 2212 nm and the average diameter of PVP nanofibers was about 84 nm. This finding is consistent with the previous work [12] reported that the combination of a high molecular weight and a low concentration of PVP being [12] electrospun at high voltage and low flow rate and the estimated entanglement of about 2 would result in the formation of beaded fibers [8].

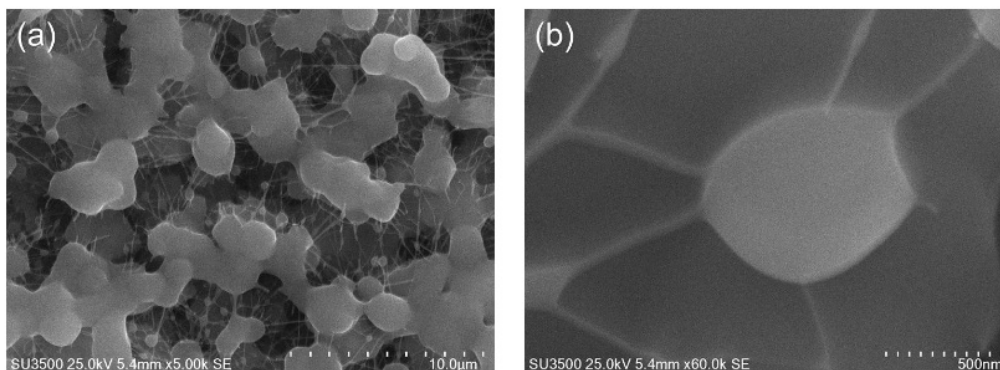


Figure 3. SEM images of PVP/GTE nanostructures magnified at (a) 5,000 and (b) 60,000 times

As given in Figure 3, the PVP/GTE nanostructures are the agglomerations of beads with nanofibers in between. It happened since the addition of GTE content reduced the concentration of PVP. When the PVP concentration was low, the PVP/GTE composite had weak core structures that caused the shell to be easily agglomerated each other [13]. The diameter distribution of PVP/GTE agglomerated beads was within the range of 231 to 1164 nm and the average diameter of PVP/GTE nanofibers was about 49 nm.

The FTIR spectra of PVP nanostructures, GTE solution, and PVP/GTE nanostructures are shown in Figure 4. The FTIR spectrum of GTE had a broad peak at 3319 cm^{-1} which are the stretching vibration of O-H group in water, alcohols, and phenols [15]. The peaks of 2944 and 2832 cm^{-1} were identified as asymmetric C-H alkanes stretch and O-H carboxyl acid stretch, respectively [15]. The group of C=C stretch in the aromatic ring and C=O stretch in polyphenols were characterized at 1655 cm^{-1} , and C-O

stretch in amino acids was detected at 1022 cm^{-1} [15]. Therefore, these findings revealed that the GTE was rich in polyphenols, carboxylic acids, and amino acids.

The FTIR spectrum of PVP had a peak at 3424 cm^{-1} which indicates O-H stretching [16]. The peaks at 2950 and 1652 cm^{-1} proved the existence of asymmetric stretching of CH_2 and stretching of C-O, respectively [16]. The C-H bending and CH_2 wagging were observed at 1423 cm^{-1} and 1288 cm^{-1} , respectively [16]. The peaks at 1018 and 568 cm^{-1} were identified as the CH_2 rock and N-C=O bending, respectively [16].

The FTIR spectrum of PVP/GTE composite resulted in the following peaks. The peak at 3396 cm^{-1} was the carboxyl group hence proving that it contained water, alcohols, and phenols [15]. Moreover, this peak could also be considered as a result of intermolecular interaction between PVP and GTE, where its position and intensity were in between those of PVP and GTE. Other phenomena happened at the wavenumbers of 1018 and 1040 cm^{-1} . In the FTIR spectrum of PVP, there was rock bending of CH_2 at 1018 cm^{-1} with low intensity. After being loaded by GTE, the peak became increasingly high and shifted to 1040 cm^{-1} , which is the stretching of C-O group in amino acid. Thus, the PVP/GTE composite had successfully been synthesized as confirmed by phenolic compounds and amino acids.

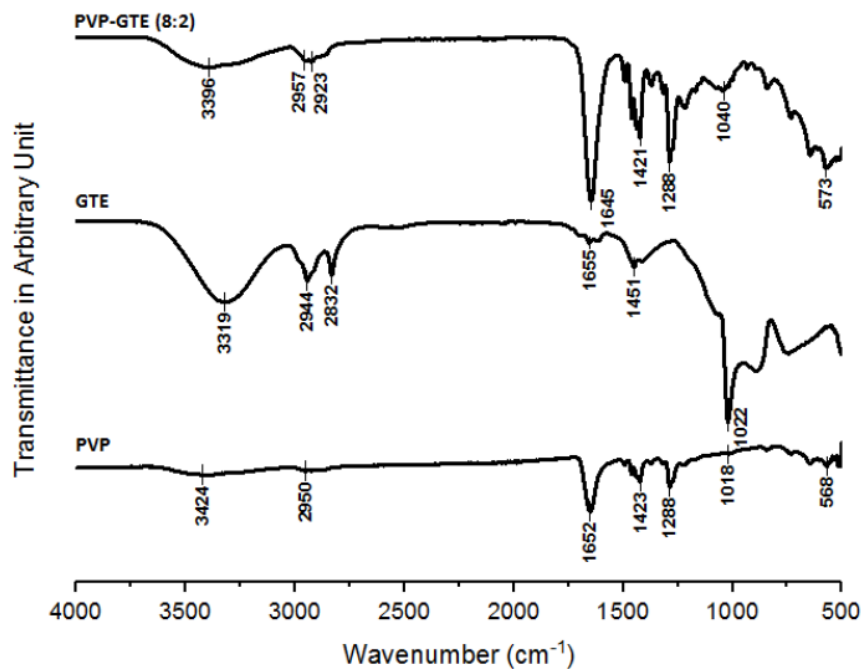


Figure 4. FTIR spectra of PVP nanostructures, GTE solution, and PVP/GTE nanostructures

4. Conclusion

The synthesis of polyvinylpyrrolidone (PVP) composite nanostructures loaded with green tea extract (GTE) as the active ingredient using electrohydrodynamic spraying technique has successfully been done. From the SEM images, it was shown that the PVP/GTE nanostructures had a combination of agglomerated beads (less spherical particles) and nanofibers. This occurred because if the PVP concentration is low, the PVP/GTE composite has weak core structures that cause the shell to be easily agglomerated each other. The intermolecular interaction between PVP and GTE in the PVP/GTE nanostructures was confirmed by the peak at 3396 cm^{-1} , which is the carboxyl group, proving that the

VP/GTE nanostructures contained water, alcohols, and phenols. Another confirmation was from the peak at 1040 cm^{-1} , which is the stretching of C-O group in amino acid.

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