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Preparation and Physicochemical Characterization of Bioplastics from Vegetable Waste/Poly (Vinyl Alcohol) and Coating with Polyeugenol

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Abstract: Plastic waste is increasingly piling up because it is not easily destroyed by rain, sunlight, or microbes that live in the soil, thereby it increases environmental damage such as soil pollution. One of the solutions to overcome the problem of using plastic is bioplastic technology. In this study, we aimed to prepare and characterize the physicochemical of bioplastics from different vegetable waste such as green spinach (GS), water spinach (WS), and moringa (M). The preparation of bioplastics was carried out with two variants: the vegetable waste as the base material/PVA and coating with polyeugenol (PEU). Physicochemical analysis of bioplastics includes surface observation by scanning electron microscopic (SEM); crystallinity by X-Ray diffraction (XRD); identification of functional groups by Fourier transform infra-red (FTIR); thermal characteristics analysis by thermogravimetric analysis (TGA), differential thermal analysis (DTA), and differential scanning calorimetric (DSC); and analysis of mineral composition by atomic absorption spectroscopy (AAS) techniques. Analysis by SEM showed that the smoothest surface was WS/PVA coating with polyeugenol (PEU) compared with other vegetable waste. The characterization results showed that the obtained bioplastic had good heat resistance up to 200 °C. XRD analysis showed that vegetable waste/PVA coated with PEU increases the crystallinity and the highest crystallinity value was M/PVA coated with PEU. The FTIR results showed the presence of C-H alkanes, C=O carboxylic acids, OH alcohols, and C-H alkenes and the spectra are similar between GS, WS, and M. Analysis by AAS showed that the bioplastic produced contained some micro and macronutrients including Ca, Cu, Fe, K, Mg, Mn, Na, and Ni and only the mineral Cu was not found in M. Thus, bioplastic from vegetable waste/PVA and coating with PEU were potentially used as mulch and packaging material.

Keywords: Bioplastic, Vegetable waste, Polyvinyl alcohol, Polyeugenol, Characterization.

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1. INTRODUCTION

Polymers are a class of chemicals that are widely used in everyday life and industry. Polymers that are widely used are plastic, rubber, fibre, and nylon (1). The use of plastic has expanded and touched almost all areas of life. Various products and equipment are made from this material because it is considered more economical, not easily broken, flexible, and lightweight. One example of a product made from plastic that is most often used by the public is plastic bags and equipment (2). Plastics have become an important necessity and their usage continues to increase. But despite these benefits, plastic is a problem for the environment.

The use of plastic as a packaging material and other equipment has caused environmental problems. Plastic waste is increasingly piling up because it is not easily destroyed by rain, sunlight, or microbes that live in the soil, thereby it can increase environmental damage such as soil pollution. One of the solutions to overcome the problem of using plastic is bioplastic technology. Bioplastics are plastics that are used like conventional plastics but are easily degraded naturally by microorganisms (3). Bioplastics can be developed by utilizing natural resources. The materials used in the manufacture of bioplastics are polymer compounds found in plants such as starch, cellulose, and lignin (4).

The utilization of food waste as a source of bioplastic feedstock is a highly sought-after result, (5) as it can significantly improve the sustainability of our economy following one of the circular economic paradigms: the substitution of plastics resulting from non-renewable sources with biodegradable bioplastics and/or derived from renewable sources (6). An example of conversion of biomass and food waste into modern biodegradable plastics are starch-based plastics, biopolymers and immerse such as polylactic acid, or materials synthesized from biomass such as PHA (7). Proteins have also recently been proposed as structural and functional biopolymers, taking advantage of their biocompatibility and various fabrication strategies (8).

Several researchers have studied the preparation and characteristics of bioplastics, including research on bioplastics from carrot, parsley, turnip, and cauliflower vegetable waste whose characteristics showed mechanical properties similar to polypropylene (PP) and starch-based bioplastics (9), mainly preparation and characterization bioplastic from starch. Among various synthetic polymers, polyvinyl alcohol (PVA) is one of the best choices because of its water solubility and biodegradability (10). On the other hand, eugenol is the main component of clove leaf oil which is mainly produced in Indonesia and polyeugenol (PEU) can be synthesised quickly, and easily, and has strong antibacterial and antioxidant activity (11).

This study aims to prepare and characterize the physicochemical properties of bioplastics derived from various vegetable wastes, including green spinach (GS), water spinach (WS), and moringa (M) added with PVA and polyeugenol (PEU). To the best of our knowledge, there have not been any reports on the preparation and characterization of physicochemical of GS, WS, and M vegetable waste added with PVA and PEU using SEM, FTIR, XRD, TGA, DSC, DTG, and AAS. This research database can be useful for designing and manufacturing bioplastic materials.

2. EXPERIMENTAL SECTION

2.1. Material and Methods

Eugenol (99.99% purity; obtained from Happy Green Co.), vegetable waste from GS, WS, M (Obtained from local market in Palu, Central Sulawesi), distilled water (Obtained from Organic Laboratory, Palu, Central Sulawesi), concentrated sulfuric acid, glacial acetic acid, methanol (p.a), ethyl acetate, n-hexane, anhydrous sodium sulfate, and PVA (all were obtained from Sigma-Aldrich).

2.2. Preparation of Bioplastic

2.2.1. Samples

One kilogram of WS waste is used as one of the samples. The samples were then properly cleaned, chopped into little pieces, and dried for two to three days. Then, using an 80-mesh screen, mix the leftover vegetable waste until it is smooth. The useful vegetable waste powder is available.

2.2.2. Synthesis of Bioplastic Films from Vegetable Waste

The glaical CH_3COOH was diluted to prepare a 1M CH_3COOH solution in water. Water vegetable powder was dissolved in a CH_3COOH solution at a concentration of 1 g of powder, and then it was vigorously stirred to hydrolyze in 20 mL of 1 M CH_3COOH solution. The vegetable dispersion was swirled on a hotplate at 70 °C for 6 hours to prevent temperature changes, then printed and allowed to cure for around 48 hours at room temperature on a Petri plate. 0.5 g of dry sample was mixed with 0.5 g of PVA in 20 mL of distilled water then stirred at 80 °C for 2 hours then cast in a petri dish, and then the solvent was evaporated at room temperature until it was dry.

2.2.3. PEU Synthesis

10 g of eugenol was weighed in a 250 mL beaker. Then 2.5 mL of H_2SO_4-CH_3COOH as a catalyst in a ratio of 4:1 (monomer: catalyst) was added gradually into the beaker while continuously stirring (room temperature, ± 5 minutes) using a magnetic stirrer. Polymer formation was indicated by the release of thick white smoke and the presence of polymer formed on the walls of the beaker. To quench polymerization, 4 drops of methanol was added onto the polymeric crude. Then the crude was left for 24 hours at room temperature until the polymer solidified and dried. Then 40 mL of ethyl acetate and 40 mL of n-hexane were added to from PEU. The mixture was then shaken until the PEU was completely dissolved. Afterwards, it was put into a 250 mL separatory funnel, then 100 mL distilled water, was then shaken and stored for 24 hours until two layers were formed. The bottom and top layers were separated and the top layer was washed for 3 times using distilled water. The top layer which is insoluble in water, PEU was dried over 1 g anhydrous Na_2SO_4 and filtered using a glass funnel. Then the filtrate was placed in a Petri dish and stored at room temperature for 48 hours.

2.2.4. Preparation of Vegetable Waste/PVA/PEU Bioplastics

Mix bioplastic with PVA in a ratio of 50%:50%, where PVA samples were weighed at 0.5 grams, and bioplastic was weighed at 0.5 grams and the mixture was redissolved in 20 mL of distilled water and stirred for 1 hour at 75 °C. Then, the mixture was poured onto a petri dish and let it sit for 24 hours at room temperature. Afterwards, polyeugenol was dissolved in 10 mL of ethyl acetate and sprayed onto the mixture with the best results from the previous treatment.

2.3. Characterization

2.3.1. Surface morphology analysis using SEM

Using a scanning electron microscope (JSM-6510LA), the bioplastics' microstructural examination was completed. After being submerged in liquid nitrogen, two distinct samples, each measuring 0.5 cm^2, of each bioplastic were randomly fractured to examine the surface of the samples. Double-sided adhesive tape was used to secure the cryo-fractured samples to the support after they had been mounted on aluminium stubs. Lastly, samples were coated with gold palladium and monitored at a working distance of 10 mm and an accelerating voltage of 10 kV.

2.3.2. Functional group analysis using FTIR
A Thermo Scientific Nicolet iS10 was used to do the FTIR measurements. One light microscope with an auto image connection and one spectrometer. The samples of bioplastics were used for these studies.

2.3.3. X-Ray Diffraction (XRD) Analysis
The bioplastic samples were observed with Rigaku-Nex QC X-ray diffractometer (XRD) at a static position using a Cu Kα radiation source (λ=1.5418 Ao) with operating conditions at 40 kV, 30 mA, angle 2θ = 10 - 60 and scan speed 2o/min.

2.3.4. Micro and Macro Mineral Analysis with AAS
The atomic absorption spectroscopy (AAS) Shimadzu was used to do mineral analysis. The samples of bioplastics were used for these studies and the mineral analysis is Ca, Cu, Fe, K, Mg, Mn, Na and Ni.

2.3.5. Thermal analysis with TGA, DTA and DSC
The thermal characteristics of TGA, DTA and DSC analyses were carried out in the Shimadzu instrument. The specimen weighed between 10 and 25 milligrams, and its temperature rose by 5 degrees Celsius every minute from 30 to 550 degrees Celsius.

3. RESULTS AND DISCUSSION

3.1. Preparation of Bioplastics from Vegetable Waste
Bioplastics were made using an environmentally friendly water-based method using 0.1M CH_3COOH and blending with PVA using water solvent. The coating with PEU is carried out using the spray method. From vegetable waste which is useless and pollutes the environment, it was made into bioplastic which is useful and environmentally friendly because it can significantly increase the sustainability of our economy following one of the circular economy paradigms: the substitution of plastic produced from non-renewable sources with bioplastic which can decompose and/or come from renewable sources. All 6 samples were made consisting of 3 samples in the form of samples with PVA added and 3 samples coated with PEU. The scheme for making bioplastic can be seen in Figure 1.

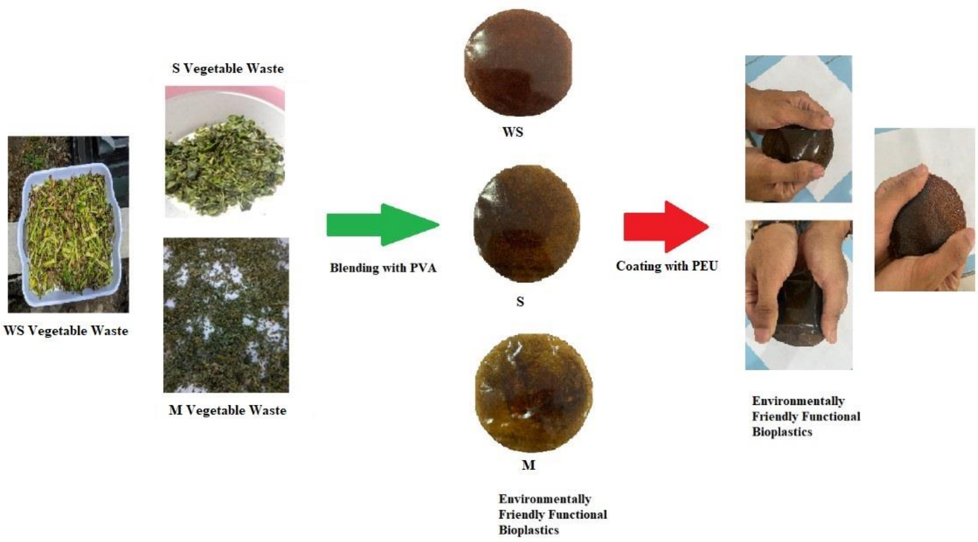

Figure 1: Preparation scheme of bioplastic from vegetable waste.

3.2. Characterization
3.2.1. Surface Morphology
To determine the surface morphology of the resulting bioplastics, analyses were carried out using SEM tecnhique. The results of the analyses can be described as follows:

The surface morphology images of the bioplastic samples were recorded with an SEM instrument with a magnification of 3000 times as shown in Figure 2. All samples showed a smooth surface, no pores, and some small parts that were less smooth. Adding PVA as a plasticizer improves compatibility, good dispersion, and a homogeneous mixture (12). Coating with PEU makes the resulting bioplastic surface smoother and more homogeneous. The less refined bioplastic part of vegetable waste is caused by an imperfect agglomeration process this can be seen from all the samples in the picture above, except for the bioplastic sample from WS waste /PVA/PEU whose surface structure is very smooth.

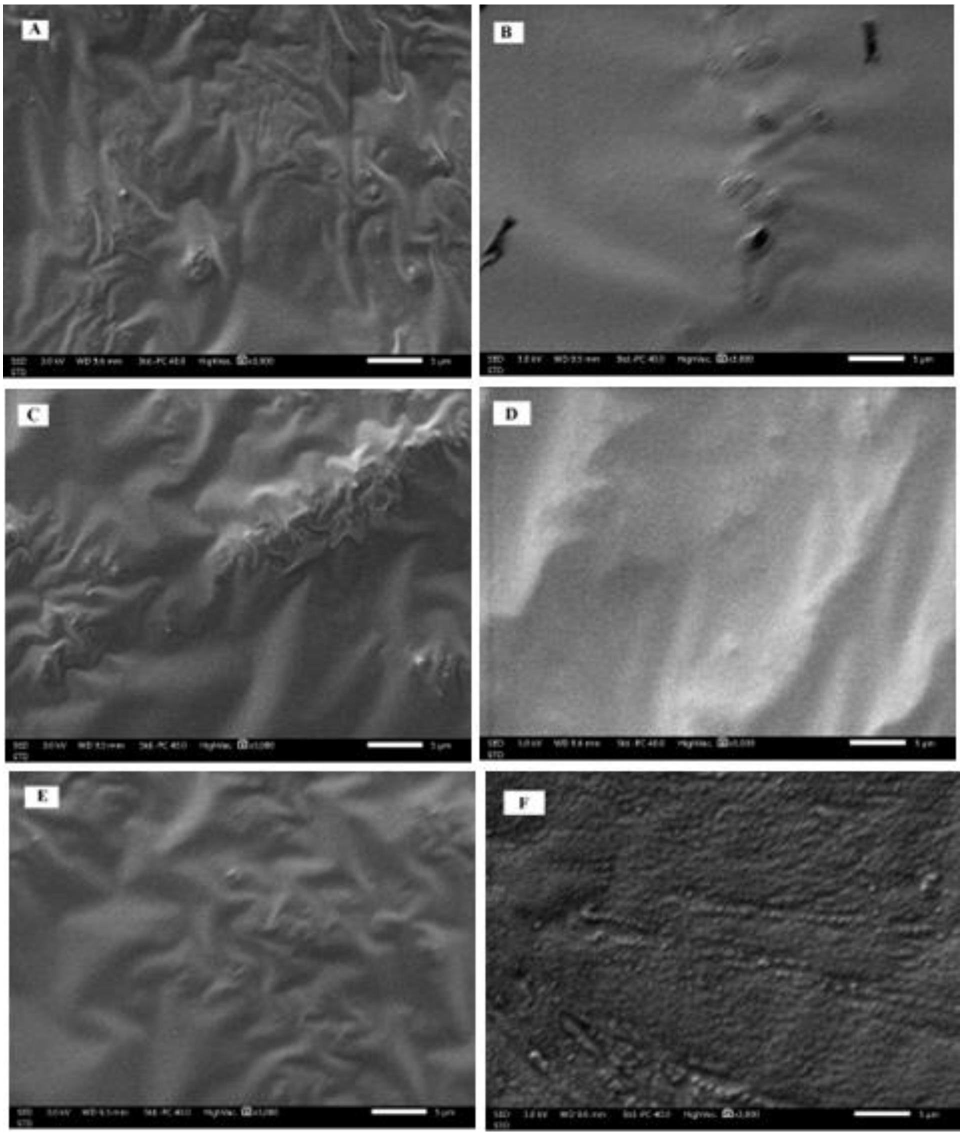

Figure 2: The SEM analyses of: A. GS/PVA, B. GS/PVA/PEU, C.WS/PVA, D.WS/PVA/PEU, E. M/PVA, F.M/PVA/PEU.

3.2.2. Bioplastic Function Group Analysis

To find out the functional groups contained in bioplastics, analysis was done using FTIR and the results of the analysis can be seen in Figure 3. The FTIR spectra analysis shows that there is a similarity between the bioplastic spectra of GS waste, WS waste, and M waste likewise with the addition of vegetable waste with PEU. The results of the FTIR analyses showed that there is absorption at 2966.52 cm^{-1} so the band that appears in the range of 2850-2970 cm^{-1} is the C-H Alkane group (13). The C-C, C-O, and COOH (carboxylic acid) bonds can be identified by the appearance of an energy band between 800 and 1300 cm^{-1} (14,15). In the results obtained, the visible group is at 1050-1300 cm^{-1}. An absorption of 1000-1300 cm^{-1} shows a typical area of the C-O ether group. The presence of the C-O ether group indicates the ability to decompose (16-18). Absorption in the 3500 cm^{-1} region indicates the presence of OH groups. The OH group is hydrophilic because water molecules can cause microorganisms in the environment to enter the bioplastic matrix thereby it damages the bioplastic (19,20). The band that appears in the range of 675-995 cm^{-1} indicates the presence of the C alkene group. Wiercigroch et al. (2017) (20) also found that the energy band at 926 cm^{-1} is a glycosidic bond in carbohydrates.

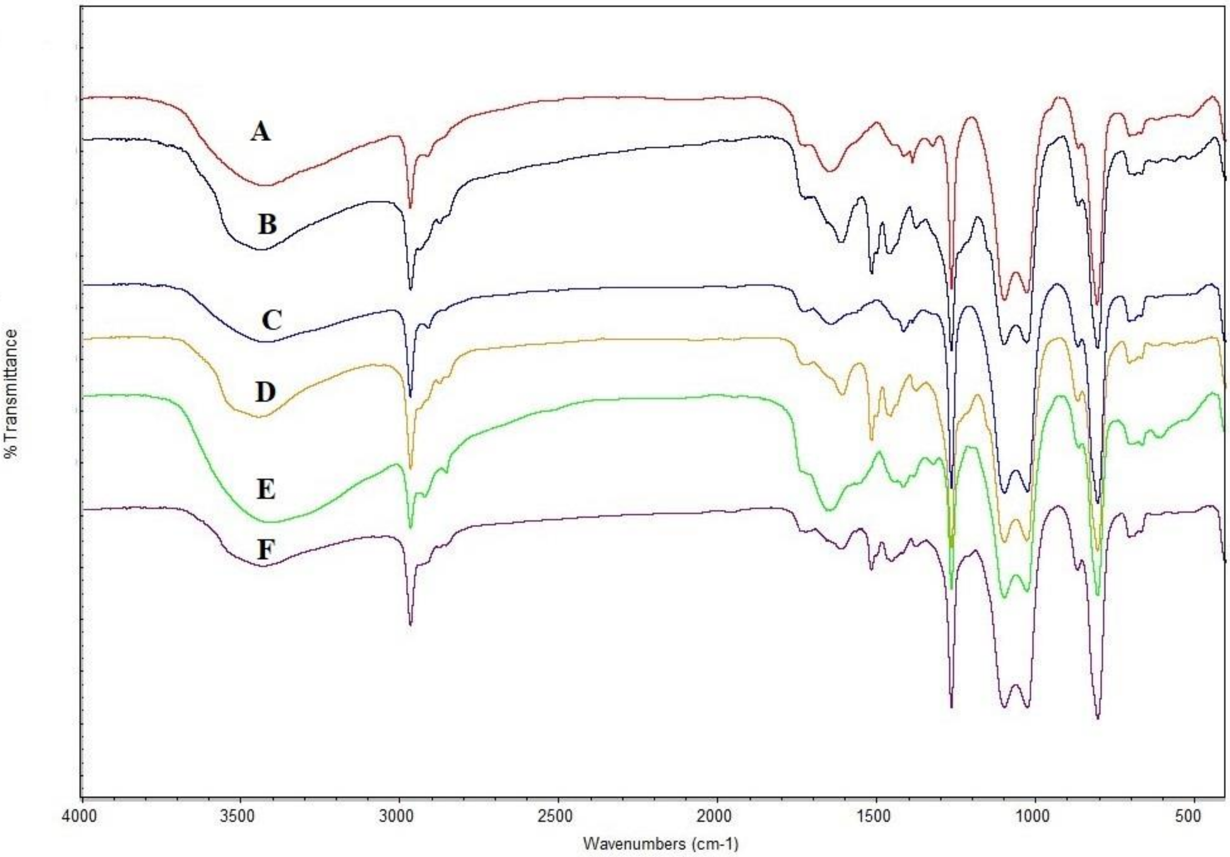

Figure 3: The FTIR spectra bioplastics sample: A. GS/PVA, B. GS/PVA/PEU, C. WS/PVA, D. WS/PVA/PEU, E. M/PVA, F.M/PVA/PEU.

3.2.3. X-Ray Diffraction (XRD) Data Analysis

The XRD spectra showed two combinations of bioplastics (vegetable waste/PVA) and (vegetable waste/PVA/PEU) (Figure 4). The diffractogram pattern of each sample has different intensities at each angle and has peaks marked by sharp curves. These peaks are known as crystal regions (21). From the diffractogram image, it can be shown that bioplastic from vegetable waste/PVA and the addition of PEU is a material that is amorphous and crystalline. The crystallinity value of bioplastics from vegetable waste/PVA increased with the addition of PEU (Table 1). Based on the results of previous research (22) using XRD, PEU shows peaks which are crystalline areas, so it is concluded that PEU has a crystalline form so that the addition of PEU increases the crystallinity of the bioplastic samples.

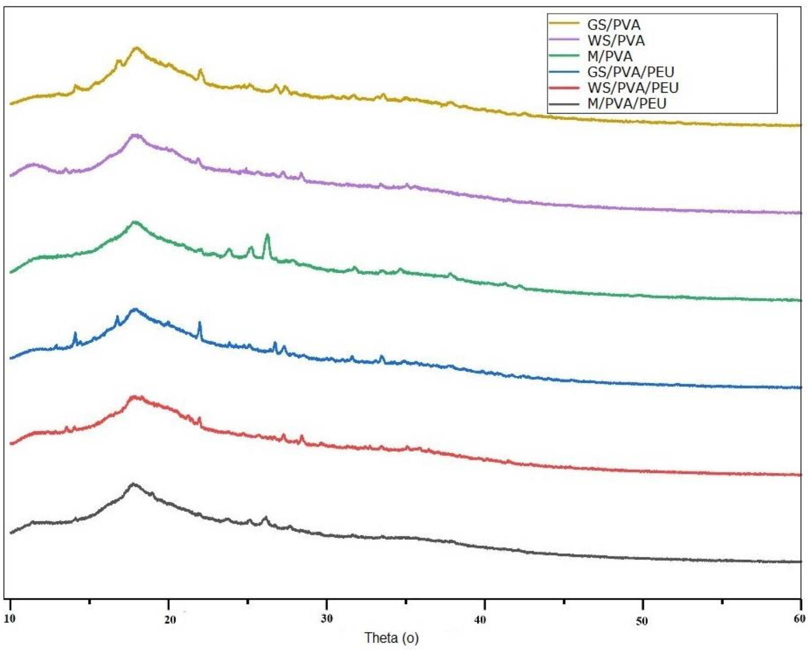

Figure 4: The XRD spectra of GS, WS and M vegetable waste/PVA and coating with PEU.

Table 1: The crystallinity of bioplastic from vegetable waste/PVA and coating with PEU.

Bioplastics	Crystallinity (%)
GS/PVA	36.2
GS/PVA/PEU	42.7
WS/PVA	39.0
WS/PVA/PEU	41.7
M/PVA	40.8
M/PVA/PEU	42.9

3.2.4. Micro and Macro Mineral Analysis with AAS

Furthermore, an analysis of the mineral composition of the selected materials was carried out using AAS to see the bioplastic potential of vegetable waste for application in fertilization. Table 2 shows that the bioplastic material developed from vegetable waste/PVA has the amount of micro and macronutrients needed for plant growth which can be used as a natural fertilizer. In this way, bioplastic can be used as film mulch which will reduce the use of inorganic fertilizers which are associated with environmental pollution and provide opportunities for the development of more sustainable agricultural systems. Vegetable waste is naturally rich in organic and mineral materials so the application of bioplastic from GS, WS, and M vegetable waste as mulch is very promising.

Table 2: The mineral compositions (mg per kg).

Bioplastics	Mineral Amount (mg per kg)							
	Ca	Cu	Fe	K	Mg	Mn	Na	Ni
GS/PVA	10600.00	5.81	202.17	2.71	4442.53	23.64	4093.69	324.62
WS/PVA	7095.36	0.09	105.33	1.51	1975.06	65.67	6933.99	50.07
M/PVA	12000.00	0	76.40	4478.64	2392.23	33.82	4879.16	118.33

3.2.5. Analysis of Bioplastics with TGA, DTA and DSC

Thermal analysis includes calorimetric differential thermal analysis (TG/DTA). TGA analysis was aimed at analyzing the loss of mass and weight due to thermal degradation. At the same time, DTA analysis aims to analyze the melting point of polymers or materials involved in endothermic exothermic processes (23). The temperature is adjustable from 31.22 to 552.35 °C. The heat of decomposition is shown in Figure 5. The results show that thermal decomposition occurs at temperatures below 155.46 °C due to the loss of light substances such as water. GS/PVA/PEU samples reduced weight by 84.68% at 552.26 °C. A 50% weight loss occurs at the temperature of 359.97 °C. The WS/PVA/PEU sample lost 92.60% at 548.97 °C and 50% weight loss at 325.79 °C. In addition, the M/PVA/PEU sample had a change of 86.20% at the temperature of 552.13 °C. GS/PVA/PEU samples were more heat stable than WS/PVA/PEU samples and M/PVA/PEU samples were more heat stable than WS/PVA/PEU samples. The breakdown of polysaccharides and the evaporation of light compounds and water occurs at this stage. Thermal properties were also analyzed by DTA (24). The results for all samples are shown in Figure 5. At this point, all samples behave on most of the same curves, with four peaks for the exothermic signal. The first endothermic peak refers to the glass transition temperature (Tg). The second endothermic peak also refers to the melting point (Tm). Samples of GS/PVA/PEU Tg 129.83 °C. The GS/PVA/PEU sample had the highest glass transition temperature of 129.83 °C. All samples had Tg between 125.73 °C and 129.83 °C. The highest Tm was obtained at 196.64 °C for the M/PVA/PEU sample. These results were comparable to those that were obtained from XRD in which the GS/PVA/PEU samples had more crystallinity than WS/PVA/PEU samples and M/PVA/PEU samples had more crystallinity than WS/PVA/PEU samples.

4. CONCLUSION

Biopolymers from vegetable waste/PVA were successfully prepared by casting from a solution and coating with PEU using the spray method. Characterization using SEM shows that the resulting bioplastic has a smooth surface and no pores. The results of FTIR analysis show the presence of C-H Alkane groups, C-C, C-O, and C-O-H bonds (carboxylic acids) and OH. Samples analyzed by XRD showed that the bioplastic produced was semicrystalline. Contains macro and micro minerals and has good heat stability. Based on the characterization results, this bioplastic has the potential to be applied as mulch and food packaging.

5. ACKNOWLEDGMENTS

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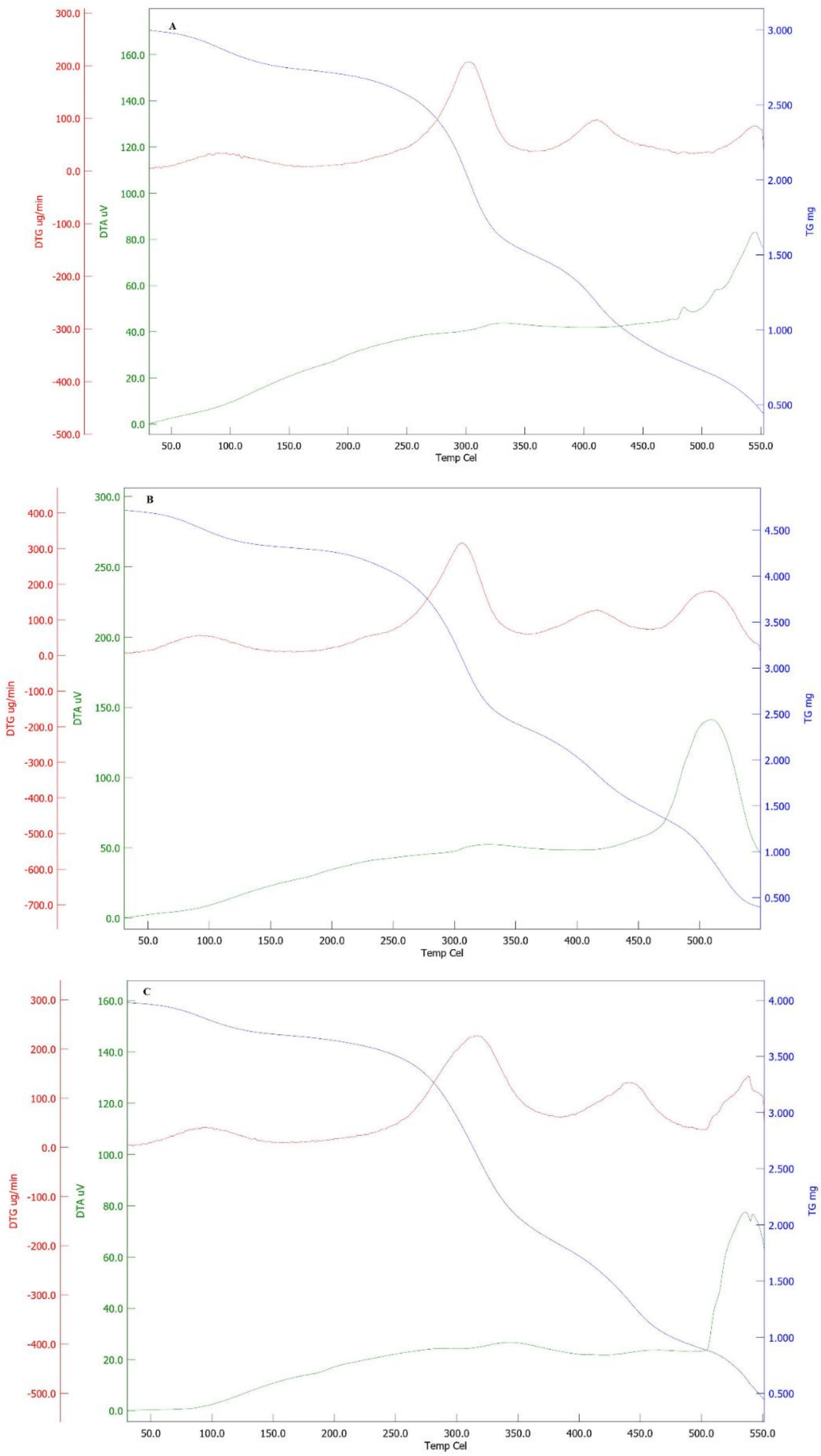

Figure 5: TGA, DTA and DSC analysis result curves for bioplastic samples from vegetable waste: A. GS (top), B. WS (middle), C. M (bottom) added with PVA and PEU.

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