



## Research article

# Preparation and characterization of polyvinyl alcohol (PVA)-glycerol composite films incorporating nanosilica from municipal solid waste incinerator bottom ash

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## ABSTRACT

This study investigates the fabrication of a composite film composed of polyvinyl alcohol (PVA) and glycerol, incorporating nanosilica derived from municipal solid waste incinerator bottom ash (BA). The nanosilica is blended with a PVA film-forming solution containing glycerol as a plasticizer. The composite films are characterized using Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), and scanning electron microscopy (SEM). Additionally, thermogravimetric analysis (TGA) is conducted to evaluate the thermal properties, while the mechanical properties are assessed in terms of tensile strength (TS) and elongation at break (EAB). The results indicate that the presence of silica nanoparticles reduces transparency and increases film thickness in the presence of glycerol. Notably, the film containing 1% silica demonstrates a significant enhancement in tensile strength, exhibiting a 50% increase compared to the film without silica. However, higher silica loadings lead to a deterioration in mechanical properties due to silica agglomeration within the polymer matrix. As expected, the presence of silica in the films slightly elevates the degradation temperature.

## 1. Introduction

Nowadays, there is growing concern over the environmental sustainability issue due to the overuse of plastic packaging film, which has impelled the advancement of biopolymer research [1]. As a result, bioplastics have drawn a lot of interest as a sustainable option for packaging applications [2]. Polyvinyl alcohol (PVA), a biopolymer, is recognized as a promising replacement for petrochemical-derived plastics, particularly in packaging contexts. PVA exhibits elevated hydrophilicity, robust compound stability, and remarkable film-forming characteristics, rendering it endowed with a plethora of advantageous attributes for deployment in packaging film [3–5]. However, PVA has some limitations including low thermal stability, and high moisture absorption when used in

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packaging [6]. The hydrogen bonding groups included in the PVA structure make it easier to combine nanocomposites with other organic and inorganic nanomaterials to enhance these properties and enrich the functionality of the polymer [7,8]. Moreover, many studies have been working to enhance the physicochemical and structural properties of the polymer, and they are particularly concerned with how the chain structure, crystallization, density, chain orientation, and packing state of polymer materials affect its penetration behaviors [9,10], the interaction between molecular size, framework, and polarity of the penetration and the material structure, which affects permeation behaviors [11,12] and the influence of polymer laminar blending on permeation behaviors [13,14]. Also, they create a variety of barrier materials and enhance the barrier qualities of materials using blocking technologies, including surface treatment [15], blends [16], nanocomposites [17], etc. In the investigation conducted by Liu et al., in 2008, they examined the PVA/titanium dioxide nanocomposite. Their findings indicated that as the concentration of carboxylated nano-TiO<sub>2</sub> increased, there was a notable reduction in the loss tangent and a substantial enhancement in the storage modulus. These effects led to a significant improvement in the tensile strength of the nanocomposites [18]. Nagalakshmi et al. (2010) proved that the thermal stability of PVA/NiO nanocomposites was enhanced with increasing NiO amount [19]. ZnO–SiO<sub>2</sub> nanocomposites were infused into PVA/CS film to enhance the shelf life of food products [20]. Salman et al. (2018) proved the capacity of PVA/nanocellulose/Ag nanocomposite films with excellent mechanical and microbiological properties, and this film can be used as antimicrobial food packaging materials [21]. Abdullah et al. (2019) prepared transparent and biodegradable water-resistant poly(vinyl alcohol)/-starch/glycerol/halloysite nanotube nanocomposite films for sustainable food packaging of lipophilic and acidic foodstuffs [22]. Moreira et al. (2020) developed an edible antimicrobial covering made of polysaccharide/PVA which is bio-active and environmentally friendly. It reduces water loss from fresh fruit storage and prevents the growth of fungi [23]. Their studies reported that composites created by combining different nanomaterials had better functional properties than pure PVA.

Among the variety of nanoparticles, silica has numerous characteristics including their biocompatibility and biodegradability, large surface areas, and abundant numbers of silanol groups. These attributes collectively contribute to the establishment of a more robust interface with polymers in composite materials, boost the efficacy of hydrogen bonding interaction and induce modifications in the crystalline behaviors within the polymeric matrix blend [24]. Therefore, in recent years, SiO<sub>2</sub> has also been frequently employed to modify polymeric materials due to its nontoxic, affordable, strong barrier, and high mechanical qualities [20,25]. SiO<sub>2</sub> has been proven to be an excellent nucleating agent for crystallization and thermal stability as well as for increasing the strength and barrier of polymers, thereby improving the general performance of composites [3,26]. Nevertheless, tetraethyl orthosilicate (TEOS) as the silica source is quite pricey. Therefore, SiO<sub>2</sub> derived from renewable resources has been the center of public concern for environmental conservation and sustainable development.

The main components of Municipal Solid Waste Incineration (MSWI) Plant ashes consist of a large amount of SiO<sub>2</sub>, therefore, it is a kind of silica source with a cheap price. Generally, utilization of the waste residues from incineration and combustion plants for production of silica-based materials is indeed viable. Several research endeavors have demonstrated success in repurposing such residues, notably through processes like the conversion of residue into zeolite via alkali-based methods [27], the synthesis of zeolitic material, and the extraction of SiO<sub>2</sub> from MSWI ash [28]. Furthermore, in a noteworthy milestone achieved in 2007, the successful synthesis of mesoporous silica materials, including MCM-41, SBA-15, and SBA-16, was accomplished using power plant bottom ash as a precursor [29]. Additionally, mesoporous silica has been prepared through a sol-gel approach employing municipal solid incineration bottom ash [30], and industrial fly ash [31]. Silica nanoparticles were synthesized from sugar industry bottom ash [32], and paper industry fly ash [33] with an alkaline extraction method. This study aims to investigate the production of silica nanoparticles through the alkaline extraction method using Bottom Ash (BA) from MSWI. The extracted SiO<sub>2</sub> nanoparticles are then incorporated into a composite film consisting of PVA and glycerol.

To our knowledge, the characteristic of PVA/glycerol film containing SiO<sub>2</sub> nanocomposite films has been less reported. Therefore, this study aimed to improve PVA packaging by incorporating nano-SiO<sub>2</sub> through the sol-gel process, utilizing glycerol as a plasticizer, to develop novel biodegradable films for packaging applications. The investigation focused on assessing the impact of SiO<sub>2</sub> concentration in the composite films on the structural properties, and mechanical, thermal, and optical characteristics of the resulting PVA/glycerol films containing SiO<sub>2</sub>.

## 2. Materials and methodologies

### 2.1. Materials

In this study, silica was extracted from the Bottom Ash (BA) which was obtained from Municipal Solid Waste Incineration Plant located at Saphan Hin, Phuket, Thailand. Poly(vinyl alcohol) was purchased from Sigma-Aldrich, Saint Louis, USA; Hydrochloric Acid (HCl) 37%; Glycerol (C<sub>3</sub>H<sub>8</sub>O<sub>3</sub>) was brought from Qrec, Newzealand; Sodium hydroxide (NaOH) 98%, Loba Chemie PVT.LTD, India.

### 2.2. Methodologies

#### 2.2.1. Extraction of silica from bottom Ash-Municipal Solid Waste Incineration Plant

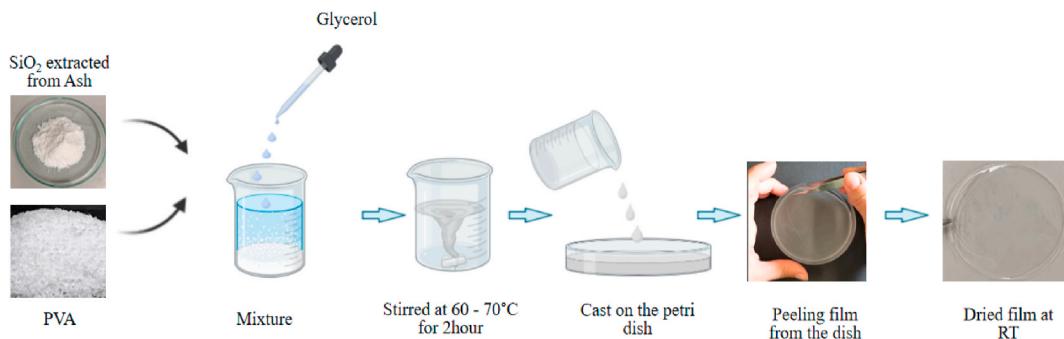
Firstly, the ingredient of BA material was examined by the X-ray Fluorescence (XRF, S2175 Ranger, Bruker, Burladingen, Germany) analysis before doing experiments, and the result was listed in Table S1.

The (BA)-municipal solid waste incineration which includes 37.5% SiO<sub>2</sub> content as the main component (Table S1 in the Supplementary material) was used as raw material for extraction of SiO<sub>2</sub>. The large granular ash was ground into small particles using a ball-mill grinder for 5 min. The resulting particles with a size less than 1 μm were collected using a mesh, and were used for silica

**Table 1**  
The film compositions.

Formulation	Glycerol (wt.%)	Silica (wt.%)
PVA-Si 0	10%	0
PVA-Si 0.5	10%	0.5%
PVA-Si 1	10%	1%
PVA-Si 3	10%	3%
PVA-Si 5	10%	5%
PVA-Si 7	10%	7%

wt.% of dry PVA.



**Fig. 1.** The diagram making the composite  $\text{SiO}_2/\text{PVA}$ -Glycerol film.

extraction with an alkaline method. The fine powder was then washed multiple times with distilled water, and dried in an oven at  $90^\circ\text{C}$  for one day.

In the round bottom flask, 5 g BA powder was stirred with 100 mL of 4 M NaOH solution, and then refluxed at  $90^\circ\text{C}$ , for 16 h. The reaction solution was then filtered through a  $0.45\ \mu\text{m}$  membrane, and the filtrate was collected for adjusting pH at 7 by using HCl 5 M and continued aging at room temperature for 24 h. The precipitated product was washed several times with hot distilled water and dried at  $80^\circ\text{C}$  for 24 h. The collected silica was analyzed again with X-ray Fluorescence (XRF, S2175 Ranger, Bruker, Burladingen, Germany) to check for impurities of Silica ( $\text{SiO}_2$  accounted for 83%), which is shown in Table S2.

### 2.2.2. Film preparation

To prepare PVA films added with Silica, 1 g PVA pellets were dispersed in 10 mL distilled water. Glycerol was employed as a plasticizing agent, and Silica powder was added into the mixture. The varied amounts of silica as the weight percentage of dry PVA are shown in Table 1. Then the mixture was stirred continually at  $60\text{--}70^\circ\text{C}$  until achieve the homogeneous solution. The PVA solution was cast on the polystyrene petri dish and dried at  $25^\circ\text{C}$  for 24h. Finally, the films were detached from the petri dish. A comparison was made between the pure PVA film, devoid of silica, and the films containing silica. The step-by-step making of the composites  $\text{SiO}_2/\text{PVA}$ -Glycerol film is shown in Fig. 1.

### 2.3. Film characterization

#### 2.3.1. Film thickness and opacity

A digital micrometer (Model) was used to measure the thickness of the film at 5 random positions of each film.

The method was used to measure the opacity of films by cutting the film into a strip ( $10\ \text{mm} \times 40\ \text{mm}$ ) and putting it along the inside wall of a cuvette. The UV-Vis spectrophotometer at 600 nm (A600) measured the absorbance. Another empty cuvette was used as a reference standard. The opacity of film ( $O$ ) was determined using the following formula (E1) [34,35].

$$O = \frac{A_{600}}{t} \quad (\text{E1})$$

where.

-  $t$  is the film's thickness (mm).

- The absorbance value at 600 nm is  $A_{600}$ .

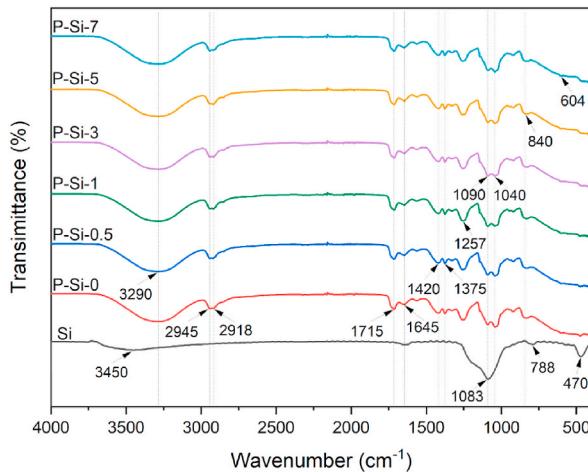
#### 2.3.2. Fourier transform infrared (FTIR) spectroscopy analysis

All films and Silica nanoparticles were analyzed with an FTIR spectrophotometer (Thermo Fisher, Nicolet iS5, USA). The sample was measured in wavelengths of 4000 to  $400\ \text{cm}^{-1}$ .

**Table 2**

The thickness, and opacity of the composite film.

Film sample	Thickness (mm)	Opacity (Abs600/nm)
PVA-Si 0	15.02 ± 0.01	0.44 ± 0.07
PVA-Si 0.5	15.20 ± 0.01	0.49 ± 0.04
PVA-Si 1	14.98 ± 0.04	0.50 ± 0.02
PVA-Si 3	14.97 ± 0.02	0.52 ± 0.41
PVA-Si 5	14.99 ± 0.03	0.53 ± 0.04
PVA-Si 7	15.07 ± 0.01	0.96 ± 0.22

**Fig. 2.** FTIR of PVA/glycerol films combined with silica nanoparticles.

### 2.3.3. X-ray diffraction (XRD) analysis

The XRD was performed on an X'Pert PRO X-ray diffractometer (X'Pert Pro MPD, Malvern Pananalytical, Bruker AXS Advance, United Kingdom) with a CuK $\alpha$  radiation source (40 kV, 40 mA, and  $\lambda = 0.15406$  nm) in the wide-angle X-ray diffractometer (WAXD) patterns. The data were captured at a scanning speed of 8°/min, in the 2θ range from 5° to 60°, with a scale factor of 0.02°.

### 2.3.4. Scanning electron microscope (SEM)

In a high vacuum and at an accelerating voltage of 12–15 kV, an SEM (Apreo, FEI, South Moravian Region, Czech Republic) was used to examine the surface and cross-section morphologies of the film. Before observation, the films were coated with gold and affixed to the specimen holder using conductive glue. Magnifications ranging from 500 to 3000x were used to visualize.

### 2.3.5. Mechanical properties

According to ASTM standard method D882-09, (Standard, 2009), tensile strength (TS) and elongation at break (EAB%) were measured at room temperature (25 °C) by using an INSTRON 5566 Machine with a tensile rate of 5 mm/min and 10 kN of tensile load. The films were conditioned in the desiccator for at least 2 days before testing. The films were cut into 100 mm × 500 mm strips and then measured at least five times for each sample, and the average of the results was calculated. Values tensile strength (TS) and elongation at break (EAB) were derived from the Instron Bluehill Universal Software (Norwood, MA, USA).

### 2.3.6. Thermal characterization

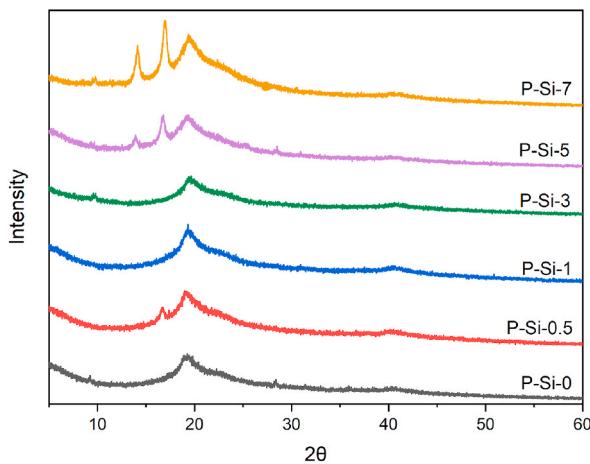
#### - Thermogravimetric analysis (TGA)

Firstly, the samples of film weighed between 7 and 10 mg and were placed in the thermal analyzer (TGA/DSC 3+, Mettler Toledo, Switzerland), which ran from 30 °C to 700 °C. Under a nitrogen environment, the TGA was conducted at a heating rate of 10 °C/min.

## 3. Results and discussion

### 3.1. Film thickness and opacity

Thickness serves as a crucial indicator for evaluating the functional properties of the film. The data reported in **Table 2** showed that the thickness value slightly increased when adding more silica content. This might be caused by the CH-based film-forming solution's



**Fig. 3.** XRD patterns of film combined silica nanoparticles and PVA/glycerol.

increased viscosity [36], leading to an impact on the resulting materials' thickness [37].

Because the appearance of the packaging film has a significant impact on customer desirability, the opacity of the materials was assessed by measuring the light transmission through the films at a wavelength of 600 nm.

**Table 2** demonstrates that the opacity of the films increased with higher silica content. The visual observation of the specimens, as depicted in the imagery, corresponded congruently with the outcomes derived from the film opacity measurements. This alignment suggested that the presence of silica had a light-inhibiting effect on the film, resulting in reduced light transmission. The observed reduction in light transmission can be attributed, at least in part, to the light-scattering phenomenon.

### 3.2. FTIR spectroscopy analysis

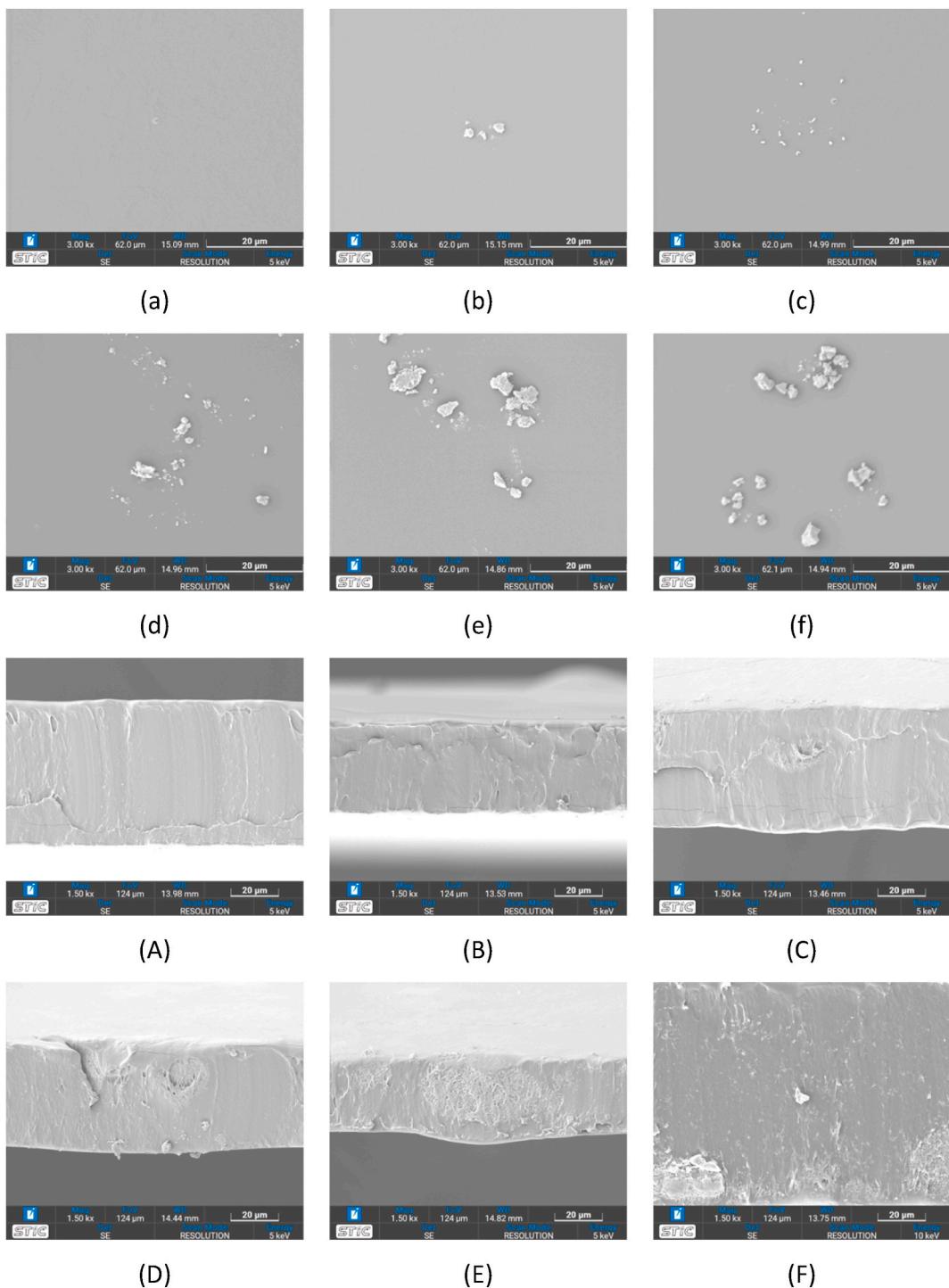
FTIR spectra were used to identify the presence of certain functional groups in the molecular structure of the composites. **Fig. 2** illustrates the FTIR spectra of PVA/Glycerol films and blending with the different percentage silica nanoparticles. For all films tested, their spectra showed  $\text{--OH}$  stretching between  $3500\text{ cm}^{-1}$  and  $3100\text{ cm}^{-1}$ ,  $\text{--CH}_2$  asymmetric stretching, and symmetrical bending at  $1945\text{ cm}^{-1}$ ,  $2918\text{ cm}^{-1}$ ,  $1420\text{ cm}^{-1}$ ,  $1357\text{ cm}^{-1}$  [38]. The peak located at around  $1040$ – $1090\text{ cm}^{-1}$  in all spectra might be associated with glycerol [38]. The stretching of  $\text{C=O}$  and  $\text{C-O}$  from acetate groups remaining in partially hydrolyzed PVA was viewed at the peak of around  $1715\text{ cm}^{-1}$  [39]. Additionally, the strong bands at  $1083\text{ cm}^{-1}$ ,  $788\text{ cm}^{-1}$ , and  $470\text{ cm}^{-1}$  were associated with the asymmetric, and symmetric  $\text{Si-O-Si}$  stretching vibration bonding [40]. The appearance of a wider peak at around  $1040$ – $1090\text{ cm}^{-1}$  in the  $\text{Si-O-Si}$  peaks also be indicative of the formation of a novel  $\text{Si-O-C}$  bond, denoting the creation of a  $\text{Si-O-C}$  linkage within the composite films through the cross-linking of silica and PVA [41].

### 3.3. XRD analysis

As shown in **Fig. 3**, all films' diffraction peaks could be seen at  $2\theta = 19.3^\circ$ , indicating PVA's semi-crystalline phase [3,21,38,42]. As the amount of silica in the sample increased, the strength of the peaks at  $2\theta = 19.3^\circ$  was slightly broader, showing that the crystalline structure of the PVA films was a little weaker. These results matched up with the mechanical tests. Even though the crystallinity drop was slight, it demonstrated that the amount of  $\text{SiO}_2$  added could have an impact on the PVA/glycerol films' crystallinity. Therefore, we can simply alter the mechanical characteristics of the PVA/glycerol biodegradable film by adjusting the  $\text{SiO}_2$  content. The absence of silica's distinctive peaks in the films further suggested that silica was evenly distributed throughout the PVA matrix as a whole [40].

### 3.4. Film structure

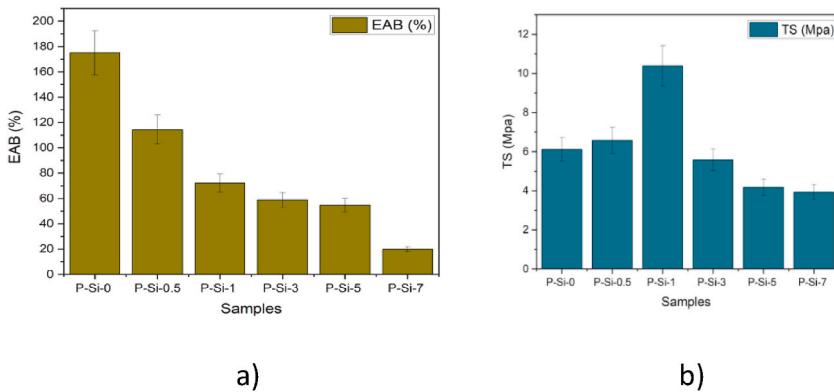
The SEM micrographs in **Fig. 4** depicted the PVA-Si composite films. Initially, the PVA-glycerol film without silica exhibited a uniform, and homogeneous surface and cross-section, as seen in **Fig. 4a** and A. However, as the silica content increased in the films (**Fig. 4b-f**), both the surface and cross-section became coarser and displayed signs of discontinuity. Furthermore, the inclusion of silica in the PVA film led to the appearance of small white spots, which became more concentrated with higher silica content. Notably, as the silica content increased from 0 to 3%, the silica particles were observed to be uniformly dispersed within the films. Nevertheless, in the PVA films containing 5% and 7% silica, some areas displayed aggregations of silica. These findings align with previous research indicating similar outcomes when  $\text{SiO}_2$  was added to PVA films beyond a certain threshold [43]. These figures serve as clear evidence that the addition of silica nanoparticles to the PVA/glycerol film resulted in a nearly uniform surface without cracks, suggesting good compatibility between silica nanoparticles and PVA/glycerol.



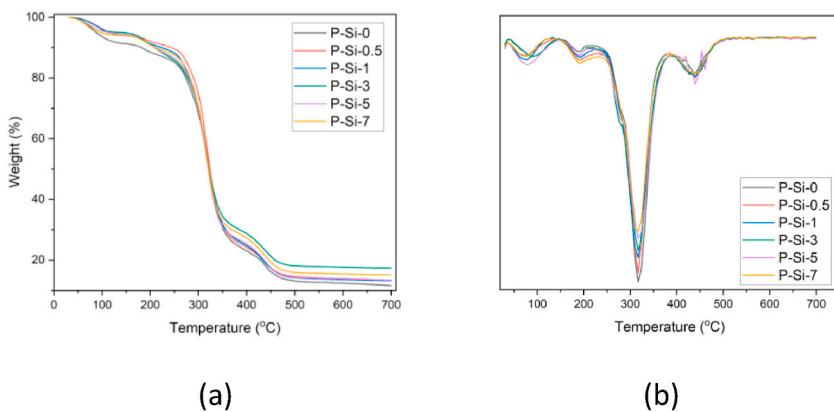
**Fig. 4.** SEM images of the surface and the cross-section of the composite films. The images correspond to different compositions: PVA-Si-0 (a) & (A); PVA-Si-0.5 (b) & (B); PVA-Si-1 (c) & (C); PVA-Si-3 (d) & (D); PVA-Si-5 (e & (E)); PVA-Si-7 (f) & (F), respectively.

### 3.5. Mechanical properties

The mechanical properties of all composite films are presented in Fig. 5. While the EAB of films declined dramatically (Fig. 5a), the TS of PVA films first climbed and subsequently significantly decreased with an increase in Silica content (above 3%). When compared to pure PVA film, the TS of the film containing 1% silica reached a maximum point and rose by over 50 % (Fig. 5b). This finding



**Fig. 5.** Mechanical analysis EAB(a) and TS(b) for PVA-Si composites films.



**Fig. 6.** TGA curves of the PVA/Si composites film.

suggests that PVA and Silica combine to produce significant interfacial contact and efficient stress transmission. It could be due to efficient hydrogen bonding interactions between Silica and carbonyl groups of PVA.

As a consequence, the films' flexibility was decreased, and their mechanical strength was enhanced. The TS was reduced for PVA composite film mixed with 3, 5, and 7 wt% silica than for PVA/Glycerol film without silica. When the percentage of Silica exceeded 1 wt%, it showed that the effects of TS were of lower quality. It may be because of the forming agglomerations when higher silica concentration, leading to the production of stress concentration spots when stretching. It would impair the functionality of the film's physical structure.

This was consistent with other outcomes obtained by adding solid additives to the polymer matrix, such as nano-SiO<sub>2</sub> and montmorillonite [1]. The starch/PVA films with 2.5% nano-SiO<sub>2</sub> added had the highest TS of all the films when the concentration of nano-SiO<sub>2</sub> was increased from 0% to 5% [44], whereas the blend films' EAB slowly dropped [45].

### 3.6. Thermal characterization

Fig. 6 displayed the TGA thermographs and derivative thermogravimetric (DTG) curves for each film. All PVA films degraded thermally in three stages, as demonstrated in Fig. 6a. The first weight loss was observed from 40 °C to 260 °C, which is when water begins to evaporate. Due to PVA's chain disintegration, a second weight loss occurred between 280 °C and 330 °C. The last stage is the decomposition of the PVA main chain and following degradation into carbon char at a range of 340 °C to 500 °C. As shown in Fig. 6b, the decomposition temperature of the PVA-Silica composite films is higher than that of pure PVA film. Additionally, when the amount of silica increased, the temperature of breakdown increased slightly. It showed that the films' thermal stability had improved as a result of the silica addition. Due to silica's effective heat insulation and gas barrier capabilities, the chain transfer process and the passage of volatile breakdown products through materials are both limited [37]. Which these properties, the biodegradable composite film is made from PVA (Polyvinyl Alcohol), silica, and glycerol which can have various practical applications due to its unique properties. This composite film can offer a combination of flexibility, mechanical strength, and barrier properties, for example - biodegradable mulch films, which can be used in agriculture as biodegradable mulch films to reduce weed growth and maintain soil moisture. It can be left in the field, gradually breaking down without harming the environment. Secondly, PVA-silica-glycerol films can be used for solar control applications to reduce heat and UV radiation transmission through windows. This composite film also can be used in

wrapping applications [46].

#### 4. Conclusion

Silica nanoparticles, extracted from BA-MSWI, with a purity of 82.56%, were employed as reinforcement in plasticized polyvinyl alcohol (PVA) films containing glycerol. The addition of silica in varying weight percentages (0.5, 1, 3, 5, and 7 wt%) resulted in notable enhancements in the mechanical and thermal stability of the composite wrapping films. As expected, the presence of silica increased and led to a slight rise in the temperature of degradation. Remarkably, the incorporation of 1 wt% silica in the films resulted in a significant improvement in tensile strength, with a 50% increase compared to silica-free samples. Conversely, the introduction of silica resulted in reduced flexibility of the wrapping films. In summary, PVA-glycerol composite films loaded with 1% silica exhibited the best performance for biodegradable wrapping applications, providing a slightly transparent appearance, enhanced tensile strength, and a sufficient level of flexibility (approximately 120%) for wrapping purposes.

#### CRediT authorship contribution statement

**Phan Thi Hong Hanh:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation, Writing – original draft. **Thitipone Suwunwong:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Resources, Validation, Visualization, Writing – review & editing. **Suchada Chantrapromma:** Data curation, Methodology, Resources, Validation, Visualization, Writing – review & editing. **Patcharanan Choto:** Investigation, Methodology, Resources, Validation, Visualization, Writing – review & editing. **Chuleeporn Thanomsilp:** Data curation, Formal analysis, Investigation, Methodology, Resources, Writing – review & editing. **Khamphe Phoungthong:** Conceptualization, Funding acquisition, Methodology, Project administration, Resources, Supervision, Validation, Writing – review & editing.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.heliyon.2024.e25963>.

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