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Incorporating novel bio-mineral materials of silicone-arrowroot modified starch for compressed powder

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Abstract

Incorporating novel bio-mineral materials of silicone-arrowroot modified starch (SA) is an environmentally friendly technique to create compressed powder, thereby integrating biodiversity with sustainability and responding to the Bio-Circular-Green (BCG) economy. The SA had improved flowability and water resistance compared with non-modified arrowroot flour (A0), concurring with the Fourier Transform Infrared (FTIR), scanning electron microscope (SEM) analyses that indicated more hydrophobic properties. To investigate the formulated dust and compressed powder, three types of binders including magnesium stearate (Ms), isopropyl myristate (Im), and mineral oil (Mo) were examined. Powder samples of all binders at each concentration gave high water resistance with floating time of more than 15 min and high compressibility at 37.88% ± 0.91% to 42.59% ± 0.28% Carr's index value and 1.61 ± 0.02 for the Hausner ratio. The hardness was 72.53 g ± 3.25 g to 98.00 g ± 3.78 g and drop test results differed depending on the density and adhesion properties of each formulation. The color stability was acceptable and not statistically significantly different when using different binders. Microbiological analysis of total bacteria count, yeast and mold in the silicone-arrowroot modified starch and pressed powder were <10 CFU·g^{-1}, while non fecal coliform, fecal coliform, *Candida albicans*, *Staphylococcus aureus*, and *Clostridium* spp. were not found.

1. Introduction

The pursuit of sustainable development has become a paramount global concern, prompting nations to align their strategies with the United Nations' Sustainable Development Goals (SDGs). The SDGs emphasize the harmonization of economic growth, environmental preservation, and social inclusivity while offering a comprehensive framework for addressing pressing challenges. In response to these imperatives, the Bio-Circular-Green (BCG) economy has emerged as a compelling approach, seeking to integrate bio-based materials, circularity, and eco-friendly practices into economic systems [1,2,3]. This research investigated whether compressed powder derived from silicone-modified arrowroot (*Tacca leontopetaloides*) starch gave enhanced functional performance, rendering it an appealing candidate for various applications, particularly those with significant environmental implications. Furthermore, the incorporation of bio-based materials epitomizes the principles of the BCG model, contributing to circularity and fostering environmentally friendly practices within economic systems [4,5,6]. The transformative potential of this local resource and its role as a catalyst for sustainable development and responsible resource management, foster a more sustainable and prosperous future in both national and global contexts.

Arrowroot is indigenous to Southeast Asia and the Pacific Islands as a tropical biennial plant that grows well in shaded conditions and has attracted significant interest owing to its diverse potential for utilization across various industries. Its starch, renowned for exceptional absorbent properties and thickening capabilities, has been used traditionally in culinary applications. However, the scope of application is limited, and the majority of cultivation is for conservation and activities in the learning center [7]. In the past, arrowroot flour was used to make a variety of sweet and savory dishes and as a home remedy for treating many diseases. However, it has now become a rare plant that is almost lost from the community.

To preserve, rehabilitate, and utilize natural resources and bio-diversity according to the green ecosystem principle, this research sought to expand the scope of utilization of arrowroot starch from only consumption [8,9] to produce a compressed powder. Traditionally, arrowroot has been used to treat acne, blemishes, freckles and nourish the skin.

The main ingredient in generally compressed powder is talcum, which is an asbestos mineral. Recent studies have shown that talc has the potential to cause cancer. The American Cancer Society (ACS) recommends using natural flour as the main ingredient to compensate for talcum and reduce the risk of health hazards [10]. Nowadays, rice flour is used as an ingredient instead of talc to reduce the risk of talc dust, and has been developed by adding natural extracts to enhance the properties of the powder, such as extracting the color from red dragon fruit together with rice flour to develop a face powder [11].

Nevertheless, natural flours have some drawbacks for use as pressed powder ingredients such as roughness and poor adhesion. Jasmine rice flour was extracted and modified using polyethylene glycol (PEG)-50 shea butter to improve physical properties in terms of flowability and water resistance, and could then stand in water for up to 5.24 min [12]. However, numerical data demonstrating the efficacy of the physical property test are scarce. Unripe banana fruit of *Musa acuminata* and *Musa sapientum* L. were extracted and 15%w/w maximum of banana starch was used for cosmetic powder. Results showed 1.27 $g \cdot g^{-1}$ to 1.53 $g \cdot g^{-1}$ water absorption capacity, poor flow properties with 31.70 to 34.84 compressibility indices, and 1.47 to 1.54 Hausner ratios, respectively [13].

Compressed powder products contain filter ingredients (such as talcum, kaolin, calcium, silica, silicates, magnesium carbonate, and metallic stearates), colors, perfume, preservatives, and binders (such as mineral oils, fatty esters, emulsifiers, lanolin, and plant-derived alternatives) [14,15,16]. The proportions of these ingredients depend on particle size and desired physical properties such as adhesive character, silkiness, smoothness, and water repellence [17]. A quality compact powder product should be strong enough to resist breakage through everyday use. Several types of binders are used in pressed powder products including dry binders (such as zinc stearate, magnesium stearate), oil binders (such as mineral oil, isopropyl myristate, lanolin derivatives), water-soluble binders (such as tragacanthin, karaya), water-repellant binders (such as mineral oil, fatty esters), and emulsion binders (such as triethanolamine stearate, glycerol) [15].

This research explored the novelty of incorporating bio-based materials using silicone-based starch modification techniques to create compressed powder formulations. Modified starch and three types of binders were investigated to assess the chemical structures, physico-chemical properties, and microbial properties of the resulting powder products. The integration of biodiversity for sustainable development is also a core aspect of this research, aligning with the principles of the BCG model which promote environmental sustainability and resource efficiency.

2. Experimental

2.1 Arrowroot flour extraction

Arrowroot tubers were collected from community enterprise in Chachoengsao province, eastern of Thailand. The starch from arrowroot tubers was extracted and modified according to previous report [18,19]. First, the arrowroot tubes were washed, peeled, rinsed twice and sliced into small pieces. Then, the cleaned arrowroot pieces were mashed with distilled water in a blender with the speed of 50,000 rpm at a ratio of 1 kg of arrowroot to 1 L of water, and the resulting suspension was filtered through a straining cloth. The filtrate was extracted four times with new drinking water. The arrowroot flour was then settled and dried in a solar incubator for 2 day to 3 day. A 95%v/v ethanol solution (cosmetic grade, Bangkok Alcohol Industrial Co.,Ltd., Bangkok, Thailand) was added to the dried arrowroot flour as the ratio 5 mL: 100 g (95%v/v ethanol : arrowroot flour) to prevent microbial effects. The mixed sample was ground again with the speed of 32,000 rpm and sieved using a 160 mesh sifter. The non-modified arrowroot flour (A0) was sealed in polypropylene plastic bags and kept at room temperature in a desiccator for use in the modification process.

2.2 Modification of arrowroot starch with silicone

To create an efficient pressed powder, arrowroot flour was modified to provide better properties in terms of homogeneous dispersion, roughness, water resistance, and skin adhesion. Silicone was chosen to enhance the hydrophobic property with a smooth and silky texture that improved long-lasting spreadability. The modification process of arrowroot starch by silicone was applied from the modification of starch in a glossiness medium [12] and the preparation of a silane/siloxane starch emulsion [20]. The silicone oil (cosmetic grade, Dow Corning Corporation, Michigan, USA) was heated at 70°C for 15 min and immediately blended with A0 powder using a grinder. The physical and chemical properties of silicone-arrowroot modified starch (SA) were examined as the raw material for pressed powder formulation.

2.3 Formulated dusting and compressed powder

Formulated dusting and compressed powder were mixed at the ratios shown in Table 1. The silicone-arrowroot modified starch (SA) and binder were mixed with the control ingredients (i.e. zinc oxide (cosmetic grade, Fukuoka Qingdao Chemicals Trading company, Qingdao, China), zinc stearate (Shanghai Canbi Pharma Ltd., Shanghai, China), calcium carbonate (food grade, Konoshima Chemical Co., Ltd., Osaka, Japan), magnesium carbonate (food grade, Konoshima Chemical Co., Ltd., Osaka, Japan), kaolin (cosmetic grade, European Performance Minerals Quality Control, Cornwall, England), allantoin (cosmetic grade, Shangdong Huasheng New Material Technology Development Co. Ltd., Shandong, China), glydant (cosmetic grade, Salicylates and Chemicals, Telangana, India), and color (Chanjao Longevity Co., Ltd., Bangkok, Thailand)) at a constant dosage in all formulations at 41 wt%. The amount of SA and binder (i.e. magnesium stearate (food grade, Faci Asia Pacific Pte Ltd., Jurang Island, Singapore), isopropyl myristate (cosmetic grade, Suriachem Sdn. Bhd., Selangor, Malaysia), and mineral oil (cosmetic grade, Maoming Zhengmao Petrochemical Co. Ltd., Guangdong, China)) in each formulation are shown in Table 1. All the powdered ingredients were mixed with the speed of 32,000 rpm for at least 1 min before the liquid ingredients were added, and the mixed samples were then ground and sieved using a 160 mesh sifter, with results shown in Table 1.

2.4 Characterization

The surface morphology of the powder samples was investigated using a scanning electron microscope (SEM); VEGA3, Tascan Orsay Holding, Czech Republic). The powder samples were coated with gold, with all images obtained using an in-lens detector at 5 kV.

Chemical structures of the raw and modified powders were analyzed using a Fourier Transform Infrared (FTIR) spectroscope (Nicolet 6700, Thermo Scientific, Madison, WI, USA), with spectra collected in the range 4000 cm^{-1} to 400 cm^{-1}. The sample was blended with KBr powder, and the mixture was compressed to form a disk. Each spectrum was scanned 64 times with a resolution of 4 cm^{-1}.

Table 1. Pressed powder formulations.

Pressed powder	Binder materials (%wt)			Silicone-arrowroot modified starch (SA) (%wt)
	Magnesium stearate (Ms)	Isopropyl myristate (Im)	Mineral oil (Mo)	
SA-Ms3	3	-	-	56
SA-Ms6	6	-	-	53
SA-Ms9	9	-	-	50
SA-Im3	-	3	-	56
SA-Im6	-	6	-	53
SA-Im9	-	9	-	50
SA-Mo3	-	-	3	56
SA-Mo6	-	-	6	53
SA-Mo9	-	-	9	50

Table 2. Scale of powder flowability [21].

Flow character	Angle of repose (deg)	Carr's index (%)	Hausner ratio
Excellent	25 - 30	< 10	1.00 - 1.11
Good	31 - 35	11 - 15	1.12 - 1.18
Fair	36 - 40	16 - 20	1.19 - 1.25
Passable	41 - 45	21 - 25	1.26 - 1.34
Poor	46 - 55	26 - 31	1.35 - 1.45
Very poor	56 - 65	32 - 37	1.46 - 1.59
Very, very poor	> 66	> 38	> 1.60

2.5 Physicochemical property of silicone-arrowroot modified starch.

2.5.1 Flowability

Powder flowability was evaluated according to the British Pharmacopeia method to determine the angle of repose. A glass funnel with an orifice 12 mm in diameter was held at a height of 60 mm above a petri dish [14]. The sample powder of 15 g was carefully poured through the funnel until a conical heap was formed on the dish [11]. The height and radius of the pile were measured. The angle of repose of the powder was calculated using Equation (1) as follows [11,12,14].

$$\text{Angle of repose } (\theta) = \tan^{-1}(h/r) \quad (1)$$

2.5.2 Compressibility index

The Carr's compressibility index was calculated directly from bulk density and tapped density using Equation (2).

$$C = \left(1 - \frac{\text{bulk density}}{\text{tapped density}}\right) \times 100 \quad (2)$$

A test sample mass (m) of 40 g was poured through a funnel into a 100 mL volumetric cylinder [11]. The volume of powder was recorded (V_0) and the bulk density was calculated using Equation (3).

$$density = \frac{m}{V} \quad (3)$$

The tapped density was obtained by lifting the cylinder at a height of 2.5 cm and then dropping it. The test was repeated until the volume of powder did not change. The volume of powder was recorded (V_T) and the tapped density was calculated.

2.5.3 Hausner ratio

The Hausner ratio (H) is related to the Carr's index (C), and indicates the powder's flowability and compressibility [14]. The Hausner ratio was calculated using Equation (4).

$$H = \frac{100}{100 - C} \quad (4)$$

A Carr's index greater than 25 is considered an indication of poor flowability/high compressibility [21]. Table 2 shows the scale of powder flowability corresponding to the compressibility index and Hausner ratio ranges.

2.5.4 Water resistance

Water resistance of the powder was obtained by weighing 5 g of the powder, pouring the substance into a beaker containing 20 mL of distilled water and then recording the floating time of each sample [11,14].

2.5.5 Moisture

The moisture content of the sample was evaluated by the Halogen Moisture Analyzer (HE53, METTLER TOLEDO, Switzerland) with 0.5 g of sample and repeated in triplicate for each sample.

2.5.6 pH

The pH of the powder was assessed by pH meter (MP220, METLER TOLEDO, Switzerland) with 10 g of powder added into a beaker of 90 mL distilled water.

2.5.7 Hardness

The hardness of the powder cake was estimated by a texture analyzer (Ta.XTplus, Stable Micro Systems, USA) with a 2 mm stainless steel needle probe and the eyeshadow test protocol. The software was used to obtain the hardness value (g) as an average of three readings per sample and repeated three times for each formulation. The higher the force required to penetrate the cake, the stronger the cake and the more effective the binder [14].

2.5.8 Drop test

A godet was dropped from a height of 30 cm onto a flat solid surface and inspected by the naked eye to assess the damage. The drop test was repeated three times for each formulation [12,14,22].

2.5.9 Color measurement

The color values of the powder samples were measured using a colorimeter (NH300, Shenzhen ThreeNH Technology Co., China). The CIELAB color space, also referred to as L*, a*, and b* color values was determined, with L* values ranging from 0 (black) to 100 (white); a* values ranging from 80 (greenness) to 100 (redness); and b* values ranging from 80 (blueness) to 70 (yellowness). All measurements were carried out in triplicate. The whiteness index (WI) was calculated using the following Equation [14].

$$\text{Whiteness Index (WI)} = 100 - \sqrt{(100 - L^*)^2 + (a^*)^2 + (b^*)^2} \quad (5)$$

Samples of all formulations were stored at room temperature for 2 months. Color measurements were investigated before and after, and the color stability (ΔC) was calculated to assess any color change.

$$\Delta C = \sqrt{(L_1^* - L_2^*)^2 + (a_1^* - a_2^*)^2 + (b_1^* - b_2^*)^2} \quad (6)$$

The tolerance level of ΔC was 2.5 [23], with lower values indicating differences not perceptible by the human eye. ΔC values over 2.5 were estimated for color degradation of the pressed powder or a noticeable difference related to the influence of binders on the color intensity of the pressed powder between samples.

2.6 Correlation for physicochemical properties

The physicochemical properties were analyzed with SPSS using the ANOVA method at the 95% significance level. Duncan's multiple range test was used to analyze the correlations between the physico-chemical properties in terms of pH, moisture, water resistance, angle of repose, compressibility index, Hausner ratio, and hardness.

2.7 Microbiological properties of arrowroot modified starch and pressed powder

The total bacteria count, yeast and mold count, non fecal coliform, fecal coliform, and *Staphylococcus aureus* were determined using the method of Sudprasert and Sansawat [18]. Ten grams of powder sample and 90 mL of 0.1% peptone solution were mixed well using a Stomacher (400 Circulator, UK) for 2 min. The mixture was diluted using the tenfold serial dilution method and 0.1 mL of sample was cultured on agar. Total bacteria count, yeast and mold count, non fecal and fecal coliform, and *Staphylococcus aureus* were determined using plate count agar (PCA), Sabouraud dextrose agar (SDA), Eosin methylene blue agar (EMB) and Mannitol salt agar (MSA), respectively. The plates were incubated at 37°C for 24 h to 48 h, except that the mold was incubated at 30°C for 7 day. Colonies were counted in the culture medium, with results reported as colony forming units ($CFU.g^{-1}$). Non fecal coliform, fecal coliform, and *Staphylococcus aureus* were assessed according to bacterial morphology and bio-chemical tests using standard bacteriological methods. *Candida albicans* analysis was performed by culturing in human serum and incubating at 37°C for 3 h [24], while *Clostridium* spp. were determined using the method of Thaitrakulpanich *et al.* [25].

3. Results and discussion

3.1 Silicone-arrowroot modified starch

The appearances of unmodified arrowroot flour (A0) and the silicone-arrowroot modified starch (SA) are presented in Figure 1. Both A0 and SA were white. The color index and whiteness index (WI) of A0 and SA are shown in Table 3, A0 had a brightness of 96.91, slightly brighter than SA at 95.97 but the whiteness index was not significantly different, ranging 95.85 to 96.70. Therefore, silicone modification had no effect on the whiteness of starch. A0 appeared as fine and diffused, while SA adhered in clusters with a more uniform and smoother appearance that exhibited a slight sheen or glossiness. SEM analysis in Figure 2 showed that A0 had a granular structure composed of oval or elliptical granules with a size of 10 μm to 25 μm, these results were similar to other research [26] with smooth [19] naturally hydrophilic surfaces [27] that absorbed water and expanded when exposed to moisture, while the SA starch granules with a size of 10 μm to 25 μm were coated with a thin layer of silicone that provided more hydrophobic properties without a change in their structure of granules. This coating caused an electrostatic interaction between the OH groups of starch with the Si-O groups of the silicone. The silicone covered the surface of the starch granules would protect against possible changes in temperature and chemical substance [28,29].

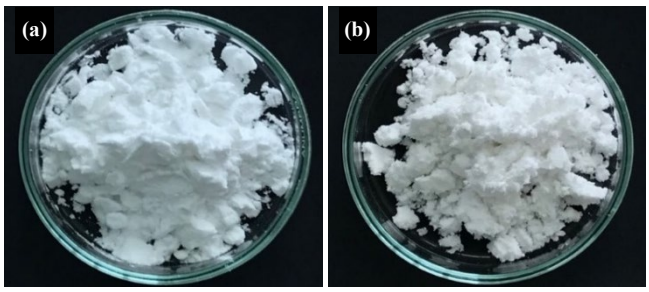

Figure 1. Appearance of (a) non-modified arrowroot flour (A0), and (b) silicone-arrowroot modified starch (SA).

Table 3. Color index and whiteness index (WI) of non-modified arrowroot flour (A0) and silicone arrowroot modified starch (SA).

Sample powder	Color index			WI
	L*	a*	b*	
A0	96.91 ± 0.06[a]	1.07 ± 0.03[a]	-0.43 ± 0.03[a]	96.70 ± 0.06[a]
SA	95.97 ± 0.09[b]	0.98 ± 0.02[a]	-0.14 ± 0.04[a]	95.85 ± 0.08[a]

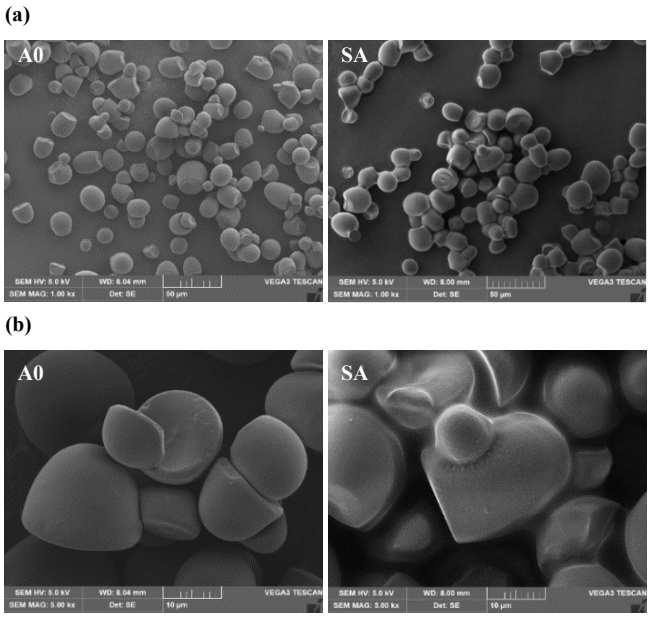

Figure 2. Scanning electron micrographs of non-modified arrowroot flour (A0) and silicone-arrowroot modified starch (SA), (a) magnified at x1000, and (b) magnified at x5000.

3.2 Chemical structure of silicone-arrowroot modified starch

The FTIR spectra of A0 and SA are presented in Figure 3. The A0 starch FTIR spectra exhibited characteristic signals related to starch structure. Broad peaks in areas of 3000 cm^{-1} to 3600 cm^{-1} and 1620 cm^{-1} were attributed to stretching vibration of hydroxyl groups (-OH). The peak at 2960 cm^{-1} related to (C-H) stretching vibrations of methylene groups ($-CH_2-$) in the starch backbone and at 994 cm^{-1} detected the vibration signal of flexion of the group (-COC-). These starch-specific signals can be characterized the arrowroot starch.

The SA modified-starch revealed the peak at 2960 cm^{-1} in the FTIR spectra corresponded to the stretching vibration of the methyl group ($-CH_3$) in the silicone. There were peaks refer to the characteristic signals of the silicone-arrowroot modified starch. Specifically, the vibration of the methyl group bonded to silicon (CH_3-Si-) and the vibration of the functional group $[-O-Si-(CH_3)_2-]_n$ was observed at 1257 $cm \cdot cm^{-1}$ and 792 $cm \cdot cm^{-1}$, respectively [30]. Therefore, arrowroot starch granules have been used to obtain modified with silicone. Peaks located at 1145 cm^{-1} and 1006 cm^{-1} were assigned to vibration of (-C-Si-O-) and (-Si-O-) in SA modified starch, respectively. Additionally, the peak at a wavenumber of 1620 cm^{-1} corresponded to the vibration of the hydroxyl group (-OH) that indicated the presence of hydroxyl groups in the modified starch. Its intensity increased due to the electrostatic interaction between the hydroxyl group of the starch and the oxygens of the silicone [29]. Consequently, it suggests a physical interaction between arrowroot starch and silicone in the modified starch.

A possible structure of the silicone-arrowroot modified starch is shown in Figure 4. During the modification process, electrostatic interactions between the OH groups of starch and the Si-O groups of silicone might occur, especially when charged groups were present in the chemical structures of both materials. Starch, as a polysaccharide, has hydroxyl groups (-OH) that are capable of carrying charges, whereas silicones, based on siloxane polymers that consist of a partial positive charge on silicon and a partial negative charge on oxygen, may include charged groups or induce charge. In addition, this electrostatic interaction is facilitated by the silicone coating on the surface of the starch granules, which generates granule stackings and it is critical in improving the hydrophobicity of the silicone-arrowroot modified starch [31,32]. Besides from protecting starch granules from external factors, the silicone coating encourages increased electrostatic interaction between starch and silicone elements. This increased interaction that is noticeable in formulations including silicone-arrowroot modified starch, has a considerable influence on the density and adhesion/cohesion forces inside the pressed powder matrix [33]. The incorporating between starch and silicone, coordinated by electrostatic forces, demonstrates the versatility and potential improvements of this composite materials across a wide range of applications, particularly in the cosmetic field.

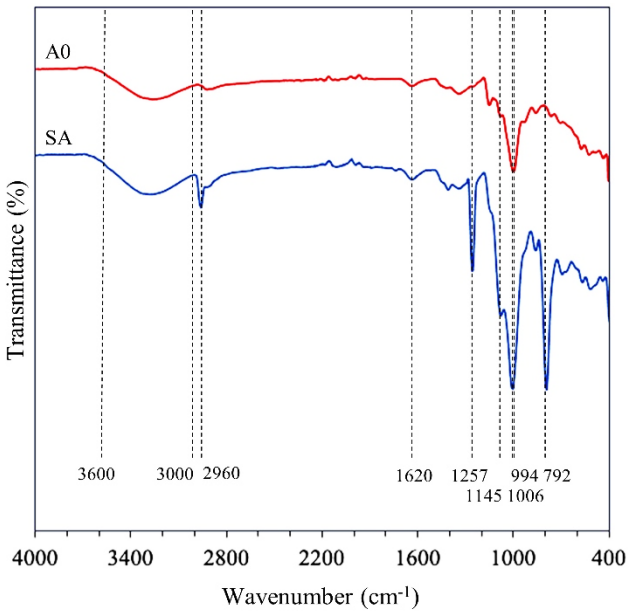

Figure 3. FTIR spectra of added arrowroot flour (A0) and silicone-arrowroot modified starch (SA).

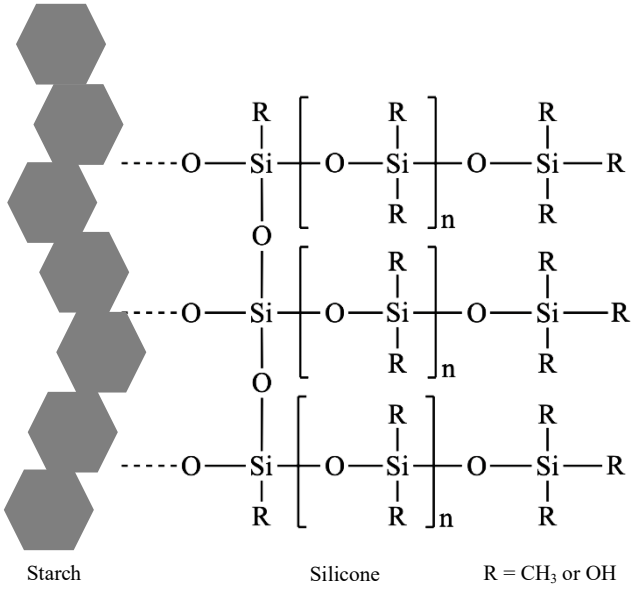

Figure 4. A possible structure of silicone-arrowroot modified starch (SA).

3.3 Physicochemical properties of silicone-arrowroot modified starch

The physicochemical parameters of SA were compared to A0 in Table 4. The pH of SA was higher than A0. Silicone compounds have more basic characteristics, causing the pH of the modified starch to rise due to chemical interactions between the starch and silicone molecules that introduced basic functional groups, thereby increasing the pH. Moisture content of the modified starch reduced and its water resistance significantly improved. This result concurred with the SEM analysis, showing that SA provided more hydrophobic properties which reduced water affinity as a result of the silicone coating on the granule surfaces [28]. SA also had a lower angle of repose and a higher Carr's index and Hausner ratio, indicating poor flowability and good compressibility. These properties were attributed to silicone-arrowroot modified starch substituting interstitial air and reducing surface tension, thereby affecting water resistance and compressibility which related to chemical structure analysis [14]. Results demonstrated that SA was a suitable material for compressed powder manufacture.

3.4 Formulated dust powders and pressed powders

The colors of the nine formulated dust powders and pressed powders, shown in Figure 5, were statistically significantly different using the ANOVA method at the 95% significance level and analyzed by Duncan's multiple range test (Table 5). When comparing each type of binder at concentrations of 3 wt%, 6 wt%, and 9 wt%, the L* values of pressed powder using magnesium stearate (Ms) as a binder increased with increasing binder concentration, whereas when using isopropyl myristate (Im) and mineral oil (Mo) as a binder, L* values decreased with increasing binder concentration. These latter two binders were in a liquid state when mixed with other compounds and the brightness or L* value decreased with increasing the binder concentration due to the reduced arrowroot starch content in the formulation. Likewise, the whiteness index (WI) of all nine formulations of pressed powder decreased with increasing binder concentration.

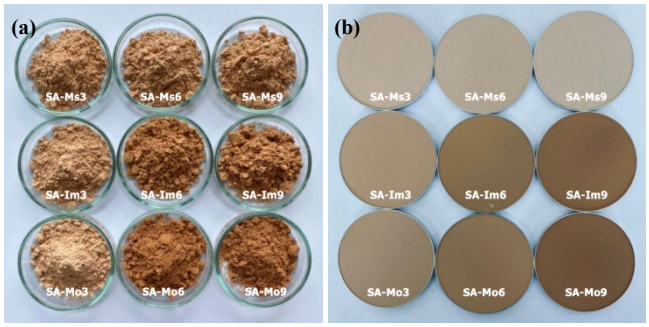

Figure 5. Photographs of (a) formulated dust powders and (b) pressed powders of SA-Ms3, SA-Ms6, SA-Ms9, SA-Im3, SA-Im6, SA-Im9, SA-Mo3, SA-Mo6, and SA-Mo9

Table 4. Properties of non-modified arrowroot flour (A0) and silicone-arrowroot modified starch (SA)

Sample powder	pH	Moisture index (wt%)	Water resistance (s)	Angle of repose (degree)	Carr's index	Hausner ratio
A0	6.32 ± 0.02^{a}	11.67 ± 0.31^{a}	34.52 ± 2.09^{a}	47.02 ± 0.84^{a}	20.58 ± 1.03^{a}	1.26 ± 0.02^{a}
SA	6.68 ± 0.06^{b}	10.71 ± 0.29^{ab}	72.83 ± 14.25^{b}	54.80 ± 1.09^{b}	48.85 ± 1.30^{b}	1.96 ± 0.05^{b}

Table 5. Color index and whiteness index (WI) values of pressed powders SA-Ms3, SA-Ms6, SA-Ms9, SA-Im3, SA-Im6, SA-Im9, SA-Mo3, SA-Mo6 and SA-Mo9.

Pressed powder formulation	Color index			WI
	L*	a*	b*	
SA-Ms3	75.97 ± 0.44^{a}	11.55 ± 0.12^{a}	16.12 ± 0.10^{a}	68.83 ± 0.43^{a}
SA-Ms6	77.51 ± 0.20^{b}	11.10 ± 0.20^{b}	15.62 ± 0.04^{b}	70.45 ± 0.18^{b}
SA-Ms9	77.85 ± 0.12^{b}	10.73 ± 0.06^{b}	15.23 ± 0.04^{b}	71.06 ± 0.10^{b}
SA-Im3	74.06 ± 1.13^{c}	12.40 ± 0.87^{c}	16.81 ± 0.87^{cd}	66.69 ± 1.62^{c}
SA-Im6	68.70 ± 1.14^{d}	$13.67 \pm 0.14d^{e}$	17.05 ± 0.19^{de}	61.82 ± 0.89^{d}
SA-Im9	64.19 ± 0.91^{e}	13.35 ± 0.16^{d}	16.17 ± 0.40^{a}	58.50 ± 0.84^{e}
SA-Mo3	74.58 ± 1.29^{c}	12.42 ± 0.74^{c}	16.91 ± 0.58^{cd}	67.03 ± 1.56^{c}
SA-Mo6	70.00 ± 0.69^{f}	14.00 ± 0.26^{e}	17.37 ± 0.21^{e}	62.61 ± 0.56^{d}
SA-Mo9	63.92 ± 0.54^{e}	13.48 ± 0.29^{d}	16.52 ± 0.54^{ac}	58.09 ± 0.41^{e}

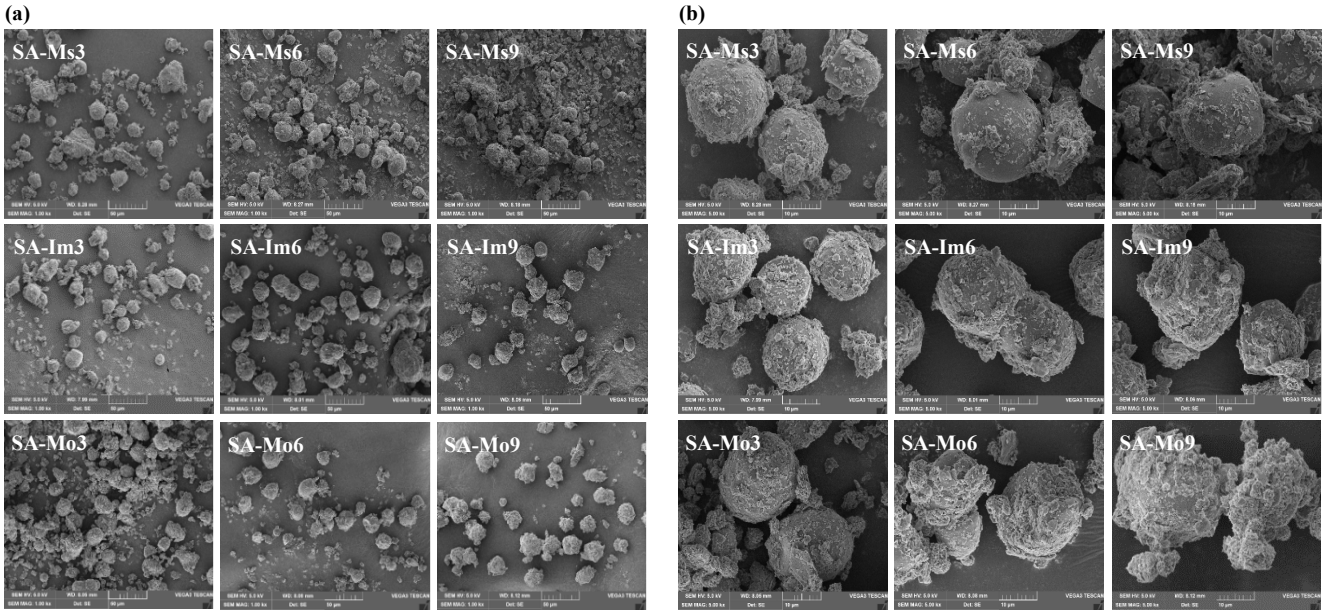

Figure 6. Scanning electron micrographs of SA-Ms3, SA-Ms6, SA-Ms9, SA-Im3, SA-Im6, SA-Im9, SA-Mo3, SA-Mo6 and SA-Mo9 (a) magnified at ×1000, and (b) magnified at ×5000.

Table 6. Properties of the formulated dust powders SA-Ms3, SA-Ms6, SA-Ms9, SA-Im3, SA-Im6, SA-Im9, SA-Mo3, SA-Mo6 and SA-Mo9.

Formulated dust powder	pH	Moisture content (%)	Angle of repose (degree)	Carr's index (%)	Hausner ratio
SA-Ms3	7.73 ± 0.10^{a}	4.89 ± 0.99^{a}	49.46 ± 0.29^{abc}	40.32 ± 1.93^{ab}	1.68 ± 0.05^{ab}
SA-Ms6	8.15 ± 0.08^{bc}	5.31 ± 0.25^{a}	49.13 ± 1.00^{ab}	37.94 ± 0.70^{a}	1.61 ± 0.02^{a}
SA-Ms9	8.2 ± 0.05^{bc}	5.07 ± 0.18^{a}	51.63 ± 0.82^{bc}	37.88 ± 0.91^{a}	1.61 ± 0.02^{a}
SA-Im3	8.11 ± 0.10^{bc}	5.95 ± 0.04^{a}	53.13 ± 1.69^{bc}	41.28 ± 0.61^{ab}	1.70 ± 0.02^{ab}
SA-Im6	8.23 ± 0.04^{bc}	5.21 ± 0.12^{a}	50.77 ± 0.37^{c}	38.23 ± 1.38^{a}	1.62 ± 0.04^{a}
SA-Im9	8.30 ± 0.05^{c}	5.25 ± 0.26^{a}	48.14 ± 0.79^{a}	42.59 ± 0.28^{bc}	1.74 ± 0.01^{bc}
SA-Mo3	7.99 ± 0.13^{b}	5.83 ± 0.05^{a}	47.64 ± 0.86^{abc}	40.29 ± 2.01^{ab}	1.68 ± 0.06^{ab}
SA-Mo6	8.19 ± 0.10^{bc}	5.84 ± 0.10^{a}	52.91 ± 1.28^{c}	40.17 ± 0.78^{ab}	1.67 ± 0.02^{ab}
SA-Mo9	8.24 ± 0.08^{bc}	5.33 ± 0.04^{a}	47.00 ± 0.45^{a}	38.99 ± 1.01^{ab}	1.64 ± 0.03^{a}

The morphology of the formulated dust powder samples was analyzed by SEM (Figure 6), which shown that the shape of the formulated dust powder samples was typically oval or elliptical-like with a size less than 30 μm. Dispersion of the particle mixtures in Figure 6(a) was larger in sample powders including Ms as a binder than in powder samples containing the other two binders because the solid Ms gave greater dispersion than the liquid binders. Micrograph of the samples (Figure 6(b)) could be observed in which the granules were covered with a thin layer of various ingredients in the formula. The powder sample used magnesium stearate (Ms), as a solid binder had a lower coating compared to the powder sample using isopropyl myristate (Im) and mineral oil (Mo), which were liquid binders. In the samples using Ms as a binder, the granules were attached with small patches of the component with a size of 0.5μm to 1 μm, and the granules still be seen as round. While the samples used Im and Mo as binders, the granules were completely coated with patches size of 1μm to 3 μm until the round shape of starch granules was no longer visible. The tetragonal or octagonal shape of patches attached to the granules' surface referred to kaolin and magnesium carbonate [34]. The coating layer would be thicker according to the concentration of the binder, which caused the powder granules of SA-Mo9 to have the largest particle size than other samples. The size and shape of elementary particles were important affecting the setting properties, coverage, transparent or opaque, and color or gloss. From the past studies, it could be found that kaolin and magnesium carbonate, which were ingredients in the formula, had rougher surfaces and were more irregular in shape, which might help to increase the setting properties, providing a semi-transparent, smooth and matt appearance [35,36].

3.5 Physicochemical properties of formulated dust powders and pressed powders

The physicochemical properties of the pressed powders were analyzed with SPSS using the ANOVA method at the 95% significance level and Duncan's multiple range test. Correlations between the physicochemical properties of formulated dust powders were determined in terms of pH, moisture content, flowability, compressibility index, and Hausner ratio, as displayed in Table 6.

The pH values of all nine dust powder formulations were between 7.73 and 8.30 (Table 6, Figure 7), showing an increase from SA with a pH of 6.68 (Table 3). Results showed that SA-Ms3 had the

lowest pH of 7.73, while SA-Im9 had the highest pH of 8.30. The pH values of the other formulations were not significantly different. The pressed powders were formulated with base components such as $CaCO_3$ and $MgCO_3$; therefore, pH values of the pressed powders were higher than the pH of SA before formulation. The pH of SA-Ms6 was not statistically different from SA-Ms9 but they were much higher than SA-Ms3, with the pH values of SA-Im3, SA-Im6, and SA-Im9 also not significantly different, along with the pH values of SA-Mo3, SA-Mo6, and SA-Mo9. Results indicated that amounts of Im, which is an oil binder, and Mo, which is a water-repellent binder, did not affect the pH values of the pressed powders. However, more than 6% of the dry binder, Ms, dramatically increased the pH of the pressed powder.

No statistically significant differences were found between the nine formulations, with results ranging from 4.89% to 5.95%. Furthermore, the water resistance of all nine dust powder formulations exceeded 15 min without the starch dissolving or sinking. Moisture contents of the formulated dust powders were lower compared to the modified starch before formulation due to the addition of insoluble compounds. Moreover, it was found that silicone-arrowroot cosmetic powder excelled PEG-50 shea butter-jasmine rice flour in terms of water resistance which was only 5.24 min [12].

The angle of repose of all nine dust powder formulations was between 47.00° and 53.13°, and considered poor flowability. The SA-Im3 formulation had the highest angle of repose at 53.13°. When comparing each type of binder, using Ms increased the angle of repose with increasing binder concentration, while the angle of repose decreased with increasing Im binder concentration and Mo gave the highest angle of repose at binder concentration of 6 wt% (SA-Mo6), as shown in Figure 7(b).

When calculating the Carr's index, all nine dust powder formulations had good compressibility. A Carr's index value greater than 25% is considered an indication of poor flowability or high compressibility. The higher the compressibility index, the more cohesive the powder, thus the more effective the binder. Table 2 shows the scale of powder flowability, corresponding compressibility and Hausner ratio ranges [1]. The Carrr's index was between 37.88% and 42.59% and the Hausner ratio was between 1.61 and 1.74. The Carr's index and Hausner ratio values of each pressed powder formulation tended to decrease with increasing binder concentration, except for formulations using Im as a binder, with the highest compressibility and Hausner ratio recorded for SA-Im9. When compared to past research of banana starch for cosmetic powder [13], the result showed the poor flow properties with 31.70 to 34.84 compressibility indices, and 1.47 to 1.54 Hausner ratios, respectively. It was found that the Carr's index and Hausner ratio values from this research had higher compressibility and suitable for development into pressed powder. The graph in Figure 7(c) displays the relationship between the compressibility of all nine formulations studied, with the Hausner ratios shown in Figure 7(d).

The physicochemical properties of the pressed powders were determined in terms of hardness immediately after pressing and 2 months after pressing, with the visual perception measure of color change presented in Table 7.

Hardness values of the nine powder formulations pressed at 1000 psi were between 72.53 g and 98.00 g, as shown in Figure 8, with hardness not significantly different between SA-Ms3, SA-Ms-6, and SA-Ms9. Using varied amounts of Ms as the binder and compressing the powder at 1000 psi had no significant effect on hardness. When using Im and Mo as the binder, SA-Im6 and SA-Mo6 gave higher hardness values than pressed powder at other binder concentrations. Magnesium stearate, as a dry binder, bonded well with particles in the pressed powder combination, even at low binder ratio of 3%, while isopropyl myristate and mineral oil, as liquid binders, required higher binder concentrations to make the particles bind together. However, at binder concentrations of 9%, the binder became a component that inhibited binding, causing the hardness of the pressed powder to decrease.

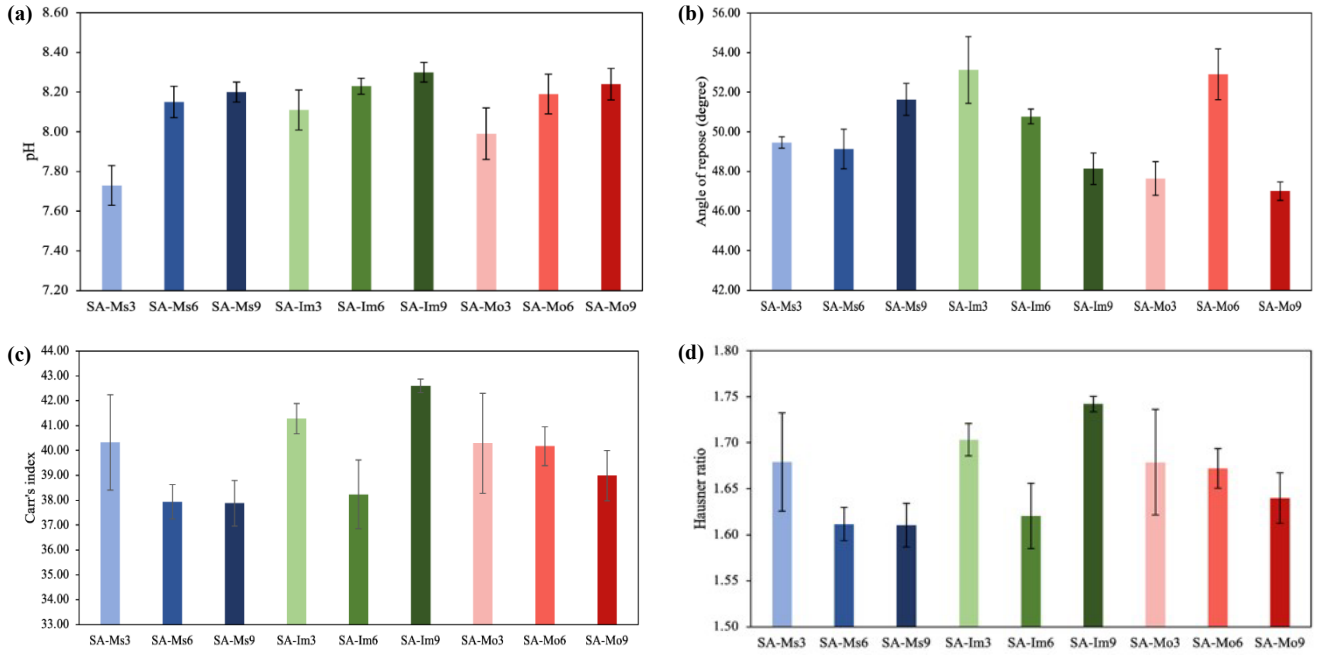

Figure 7. Physicochemical properties; (a) pH, (b) angle of repose, (c) compressibility index, and (d) Hausner ratio of powder samples; SA-Ms3, SA-Ms6, SA-Ms9, SA-Im3, SA-Im6, SA-Im9, SA-Mo3, SA-Mo6, and SA-Mo9.

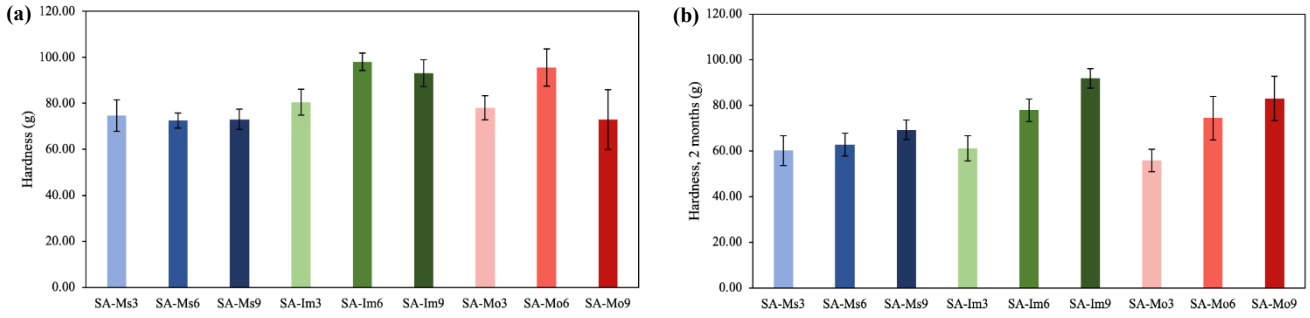

Figure 8. Hardness of the pressed powders SA-Ms3, SA-Ms6, SA-Ms9, SA-Im3, SA-Im6, SA-Im9, SA-Mo3, SA-Mo6 and SA-Mo9: (a) immediately after pressing, and (b) after 2 months.

Table 7. Hardness properties of the pressed powder formulations SA-Ms3, SA-Ms6, SA-Ms9, SA-Im3, SA-Im6, SA-Im9, SA-Mo3, SA-Mo6 and SA-Mo9 initially and after 2 months.

Pressed powder formulation	Hardness (g)	Hardness after 2 months (g)	ΔC
SA-Ms3	74.67 ± 6.91^{ab}	60.18 ± 6.56^{ab}	0.66 ± 0.11^{a}
SA-Ms6	72.53 ± 3.25^{a}	62.86 ± 5.01^{ab}	0.67 ± 0.10^{a}
SA-Ms9	72.99 ± 4.44^{a}	69.28 ± 4.27^{abc}	1.87 ± 1.00^{a}
SA-Im3	80.49 ± 5.63^{abc}	61.27 ± 5.49^{ab}	0.67 ± 0.04^{a}
SA-Im6	98.00 ± 3.78^{c}	77.85 ± 4.90^{bcd}	0.79 ± 0.10^{a}
SA-Im9	93.04 ± 5.82^{abc}	91.77 ± 4.22^{d}	1.20 ± 0.16^{a}
SA-Mo3	78.06 ± 5.2^{abc}	55.89 ± 4.96^{a}	0.84 ± 0.06^{a}
SA-Mo6	95.53 ± 8.10^{bc}	74.44 ± 9.49^{abcd}	0.96 ± 0.23^{a}
SA-Mo9	72.89 ± 12.97^{a}	83.02 ± 9.65^{cd}	1.37 ± 0.31^{a}

Table 8. Integrity of the pressed powders after 1, 2 and 3 drop tests.

Pressed powder formulation	Drop tests		
	1st Drop	2nd Drop	3rd Drop
SA-Ms3	○	–	×
SA-Ms6	○	○	–
SA-Ms9	○	○	–
SA-Im3	○	○	×
SA-Im6	○	○	○
SA-Im9	○	○	○
SA-Mo3	○	–	×
SA-Mo6	○	○	–
SA-Mo9	○	–	×

After storing the pressed powder at room temperature for 2 months, the hardness values increased with increasing amounts of each binder, as shown in Figure 8(b). Pressed powders using Im as the binder gave the highest hardness at all binder concentrations. When using Ms as the binder, the hardness was similar at each concentration, while for pressed powder using Mo as the binder, hardness differed at each concentration.

Color measurements of the pressed powders after storing at room temperature for 2 months were investigated by calculating the color stability, ΔC. Results showed that the tolerance level of ΔC was below 2.5, indicating that color variations of the pressed powders were too small for the human eye to perceive. The color stability of each formulation was acceptable and not statistically significantly different.

Table 8 shows the results of the drop test, with ○ indicating unacceptable damage, i.e., the pressed powder broke or cracked; – indicating minor damage, i.e., the pressed powder showed some chipping; and × indicating no damage. In general, addition of all binders increased the elasticity of the pressed powder proportionally with increased binder concentration. The experimental results showed that SA-Im6, SA-Im9, and SA-Mo9 were the three formulations that did not crack after three drops, while SA-Ms6, SA-Ms9, and SA-Mo6 showed minor damage after the third drop, SA-Im3 showed unacceptable damage after the third drop and SA-Ms3 and SA-Mo3 showed minor damage after the 2nd drop and unacceptable damage after the 3rd drop. These results indicated that the three types of binders at 3% binder concentration were unable to bind the powder sufficiently to be durable enough to withstand impact under the 1000 psi pressure condition.

Table 9. Microbiological properties of silicone-arrowroot modified starch (SA) and pressed powder.

Microbiological analysis	Microorganisms ($CFU.g^{-1}$)	
	Si-arrowroot modified starch	Pressed powder
Total bacteria count	<10	<10
Yeast	<10	<10
Mold	<10	<10
Non fecal coliform	-	-
Fecal coliform	-	-
Candida albicans	-	-
Staphylococcus aureus	-	-
Clostridium spp.	-	-

3.6 Microbiological properties of compressed powders

Microbiological analysis of total bacteria count, yeast and mold in the silicone-arrowroot modified starch (SA) and pressed powder were counted at <10 $CFU.g^{-1}$, while non fecal coliform, fecal coliform, *Candida albicans*, *Staphylococcus aureus*, and *Clostridium* spp. were not found (Table 9). All parameters were within the range of Thai Community Product Standard 910/2560 of pressed powder. SA and pressed powder were added many ingredients such as zinc oxide, zinc stearate, glydant which were inhibit microbial. It was reported that some of these cosmetic powders were contaminated with spores of microbial and could support their growth when they were poorly preserved. Microbial contaminants in unused cosmetic powder were common because of the environment in which the powders were manufactured, packed and the ingredients themselves [37].

4. Conclusions

Silicone-arrowroot modified starch (SA) using 10% silicone was successfully developed and provided improved physical properties in terms of flowability and water resistance compared with the non-modified arrowroot flour (A0). The FTIR spectra results indicated that modified starch replaced the interstitial air and reduced surface tension by increasing intermolecular interaction between the starch molecules, leading to increased density and adhesion/cohesion forces, while SEM analysis showed that the SA granules were coated with a thin layer of silicone that increase the hydrophobic properties.

The physicochemical test results of the formulated dust powders showed that all binders at each concentration gave high compressibility properties. When the powder formulations were pressed at 1000 psi, the hardness values and drop test results differed depending on the density and adhesion properties of each formulation. Color measurements also showed that increasing binder concentration provided a more intense color profile, while color stability was acceptable and not statistically significantly different. Microbiological analysis of total bacteria count, yeast and mold in SA and pressed powder were <10 $CFU.g^{-1}$, while non fecal coliform, fecal coliform, *Candida albicans*, *Staphylococcus aureus*, and *Clostridium* spp. were not found.

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