

Effect of rice variety on the physicochemical properties of the modified rice powders and their derived mucoadhesive gels

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Summary

In the present study; the glutinous Niaw Sanpatong (NSP) and Niaw Koko-6 (NKK), and the non-glutinous Jasmine (JM) and Saohai (SH) were chemically modified. The difference of these rice varieties on the physicochemical characteristics of the modified rice powders and the properties of the derived gels were evaluated. X-ray diffractometer was used for crystalline structure investigation of the rice powders and gels. A parallel plate rheometer was used to measure the rheological property of the gels. It was found that the non-glutinous varieties produced gels with higher mucoadhesive properties than the glutinous rice. Rheological behavior of JM and SH gels was pseudoplastic without yield value whereas that of NSP and NKK gels was plastic with the yield values of 1077.4 ± 185.9 and 536.1 ± 45.8 millipascals-second (mPas), respectively. These different properties are considered to be due to the amylose content in different rice variety. The results suggest that the non-glutinous rice varieties with high amylose content are the most suitable for preparing gels as local delivery systems *via* the mucosal membrane.

Keywords: Rice variety, rice gel, biodegradable, mucoadhesive, amylose content

1. Introduction

Rice (*Oryza sativa* L.) is the principal staple food for half the world's population. Rice grain is an important raw material for rice starch and nutrients like lipids and proteins. Moreover, rice bran is a rich source of bioactive antioxidant compounds (1). Rice powder is usually produced by milling whole rice grains including rice bran. Therefore, biodegradable products derived from rice powder are composed of various nutrients with antioxidant components.

Local delivery of drug *via* buccal, nasal, or vaginal mucosal membrane is receiving increased attention as for avoidance of acid hydrolysis in the gastrointestinal tract and hepatic first-pass effects (2). Drug delivery *via* mucosal membrane can be used for local therapy (3,4). Among several kinds of trans-mucosal dosage

forms, gel is one of the most preferable because of its excellent mucoadhesiveness, comfort, and easy dispersion throughout the mucosa. Traditional gel bases are made of synthetic polymers, thus causing serious environmental problems. Gel bases derived from rice powder are of interest because of the advantages of rice mentioned above. However, rice varieties might affect the characteristics of the derived gels. Previous studies reported that there are great variations in the amylose content in the rice starch of different varieties (5). The amylose variation was reported to influence the properties of the films derived from rice (6,7). Therefore, it is necessary to study the relevant properties of various rice varieties to choose the one most suitable for formulation of appropriated gel bases as mucosal delivery systems. To our knowledge, there have been no previous studies examining the physicochemical characteristics of rice varieties which may affect the properties of the derived gels particularly on their rheological and mucoadhesive properties. The aim of this study was to investigate the effect of rice variety on the physicochemical properties of the modified rice powders and their derived mucoadhesive gels.

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2. Materials and Methods

2.1. Rice materials and chemicals

Milled rice grains of four common varieties in Thailand; Jasmine (JM), Saohai (SH), Niaw Sanpatong (NSP) and Niaw Koko-6 (NKK) harvested during April – September 2013 were used. These rice varieties are the most popular rice in Southeast Asian countries, particularly in Thailand. JM is classified as aromatic non-glutinous rice with pleasant odor. SH, NSP and NKK are the odorless rice varieties. NSP and NKK are classified as glutinous whereas SH is non-glutinous rice. Silver nitrate and monochloroacetic acid were obtained from Sigma Chemical Co. (St. Louis, MO, USA). Methanol and glacial acetic acid were from RCI Lab-scan Co., Ltd. (Bangkok, Thailand). All other chemicals and solvents were of AR grade or the highest grade available.

2.2. Composition analysis of rice

The composition analysis of the raw rice powders was performed according to the AOAC guidance (8). Kjeldahl nitrogen method was used for determining crude protein content with the nitrogen-to-protein conversion factor of 6.25. Soxhlet extraction method was used for fat content determination with petroleum ether as a solvent using a BUCHI Soxlet fat extraction unit over a 2 h period. Dry incineration in a muffle furnace at 550°C for 24 h was used to determine the ash content. Carbohydrates content was approximately determined by subtraction from the contents of the others components.

2.3. Determination of amylose and moisture content

Raw rice powder of each variety prepared by wet milling method previously described by Okonogi *et al.* (7), were analyzed for amylose content according to the method described by Juliano (9). Moisture content of the raw rice powder was determined using a Kett F-IA moisture content balance with halogen heating. The heating temperature was set at 105°C. The exact weight of sample before and after being heated to a constant weight was recorded. The moisture content of the samples was calculated on a wet basis.

2.4. Modification of rice

Rice modification was done according to the previous method described by Okonogi *et al.* (7) with some modification. Briefly, the raw rice powder was subjected to etherification using methanol-water mixture as a medium. The reaction was carried out in a 500-mL three necked round-bottom flask, equipped with motor-driven stirrer. A 50% sodium hydroxide aqueous solution was firstly mixed with methanol at a

weight ratio of 1:4. After adding raw rice powder, the mixture was stirred until homogenous, and then proper amount of monochloroacetic acid was added. The mixture was stirred at 60°C for 3 h. The solid granules obtained were collected. The solid phase was washed several times with 95% ethanol until the silver nitrate test for chloride of the filtrate was negative. The dried solid product was pulverized. The modified rice powder that passed 80-mesh sieve was used for further studies.

2.5. Morphology and internal solid structure of rice powders

The morphology of rice particles were investigated by scanning electron microscope (SEM) using a JEOL JSM-5410LV (Japan) equipped with a large field detector. The acceleration voltage was 10-20 kV under low vacuum mode (0.7-0.8 torr). The internal solid structure of the obtained rice powders was characterized by means of X-ray diffraction (XRD) using a Siemens D-500 X-ray diffractometer with Cu K α radiation at a voltage of 30 kV and 15 mA. The rice samples were scanned between $2\theta = 5-60^\circ$ with a scanning speed of 5°/min. Prior to testing, the rice samples were dried at 50°C for 24 h and stored in a desiccator.

2.6. Solubility index

The solubility index of the rice powders was evaluated using the method previously described (10) with some modification. Rice powders (1 g) were gradually dispersed at room temperature in 250 mL of water by stirring at 100 rpm. After 1 h the solutions were filtered through Whatman (No. 1) filter papers and the weighed portions of the filtrate were dried at 60°C under vacuum until a constant weight was reached. The solubilization index was expressed as the percentage ratios of the solubilized rice/initial weighed rice. The higher of the index value, the higher the aqueous solubility of the samples.

2.7. Gel preparation

The modified rice powders were weighed and dispersed in distilled water to obtain 10% w/w rice dispersion. The dispersions were heated to 90°C in a closed chamber for 2 h and gently stirred to obtain homogenous gels without air bubble formation. The physical appearance of the gels was observed visually.

2.8. Internal solid structure of gels

The internal solid structure of the gels was investigated by using a Siemens D-500 X-ray diffractometer according to the procedures described in 2.4.1. The freshly prepared gel sample was studied without any prior treatment.

2.9. Rheological behavior of gels

The gels were investigated for their rheological behavior using a Brookfield rheometer R/S-CPS (USA) with a parallel plate and plate gap of 1000 μm . The gel sample was gently loaded onto the rheometer plate using a microspatula. Care was taken to minimize shearing during sample removal and sample loading. The rheological behavior of the samples was characterized over a range of 0-1000 s^{-1} for a period of 3 min. All studies were done at $30 \pm 2^\circ\text{C}$. The measurements were made in triplicate. The stress-shearing rate profile of any gel was obtained from a plot of shear stress versus shear rate under the given experimental conditions. The rheological parameters of the samples such as average apparent viscosity and yield value were calculated by using Rheo3000 program.

2.10. Mucoadhesive study

The gels were examined for mucoadhesive property using an *ex vivo* mucoadhesive test previously described (11) with some modification. Gels with the exact weight of 1 g were applied homogeneously on a 2 cm \times 2 cm area of a porcine intestinal mucosal membrane. After that the membrane was fixed on the internal side of a beaker which was kept in a bath at $37 \pm 1^\circ\text{C}$. The beaker was filled with 800 mL water at the same temperature. A 150 rpm stirring rate was subsequently applied to simulate the *in vivo* cavity environment to which the gels will be subjected. The time of gel detachment or disappearance from the membrane was recorded.

2.11. Statistical analysis

Descriptive statistics for continuous variables were calculated and reported as a mean \pm standard deviation. Data were analyzed using a one-way analysis of variance and Duncan's multiple range test ($p < 0.05$) using SPSS software version 11.

3. Results and Discussion

3.1. Composition analysis

The content of chemical compounds defines the nutritional quality of rice. It is well known that the major nutritional component of rice grain is carbohydrate at approximately 70-90% or more. This difference is dependent on many factors including the variety of rice. It was previously reported that total carbohydrate content in the brown rice grains of six varieties grown in the Philippines was between 72-82% (5). Starches existing in different rice varieties differ in stability. Sagum and Arcot (12) reported that three raw rice varieties had similar total carbohydrate content of 82-83% w/w. The four native rice varieties used in the present study contained

Table 1. Composition of the raw rice powders

Composition (%)	Rice Varieties			
	Non-glutinous rice		Glutinous rice	
	JM	SH	NSP	NKK
Carbohydrates	92.0 \pm 0.8	92.1 \pm 0.2	91.5 \pm 0.8	91.9 \pm 0.3
Lipids	0.5 \pm 0.3	0.4 \pm 0.3	0.7 \pm 0.3	0.6 \pm 0.1
Proteins	7.2 \pm 0.4	7.2 \pm 0.1	7.4 \pm 0.2	7.3 \pm 0.5
Ash	0.3 \pm 0.1	0.3 \pm 0.1	0.2 \pm 0.1	0.2 \pm 0.1

carbohydrate content of approximately 91-92% (Table 1), significantly higher than that previously reported. The non-glutinous rice showed slightly higher carbohydrate content than the glutinous varieties. The glutinous varieties had slightly higher protein and lipid but less ash than the non-glutinous rice. Our results of protein content of the glutinous rice was consistent with Keeratipibul *et al.* (13), who worked with similar Thai glutinous rice varieties and reported their protein contents of 7.0-7.5%. However, Thumrongchote *et al.* (14), who worked with different non-glutinous varieties, reported their protein content slightly less than our results. This was considered to be due to the difference in rice varieties. Fat content for all the rice varieties evaluated ranged between 0.4-0.7% which is in good agreement with other previous reports, while ash content of our white rice samples ranged between 0.2-0.4% which significantly lower than that of the black rice varieties reported previously (15).

Moisture content and moisture migration can cause physical, chemical and biological changes in materials (16). According to the definition, moisture content is expressed on a wet basis or a dry basis. In this study, the moisture content of the rice samples was calculated as the wet basis. Among the four rice varieties, NKK showed the lowest moisture content of 2.6% whereas SH showed the highest value of 7.2%. The moisture contents of JM and NSP were similarly moderate level at 3.4 and 4.3%, respectively. Moisture content of raw rice influences its storage properties as it is related to the water activity of microorganisms (17). The results in the present study suggest that attention should be paid to the storage of rice with high moisture content such as SH because of the high risk of microorganism contamination.

The amylose content among the four rice varieties was significantly different. SH had the highest amylose content of $21.8 \pm 0.3\%$ followed closely by JM ($17.5 \pm 0.5\%$) whereas NSP presented the lowest amount of $4.0 \pm 0.5\%$ followed by NKK ($7.4 \pm 0.4\%$). It was noted that the amylose content in the non-glutinous rice was significantly higher than that of the glutinous rice. These quantity differences of rice compositions mutually influenced the characteristics of their respective modified rice powders and gels.

3.2. Modification of rice

Preferable hydrophilic gels can be formed by using gelling agents with high ability to dissolve in water.

However, raw rice starch has low water solubility. Therefore, modification of rice starch based on physical, chemical, and biological reactions has been suggested to solve this problem (18). In the present study, chemical modification based on carboxymethylated etherification was used because the modified starches obtained from this etherification reaction were reported to cover a wide range of applications including pharmaceutical and biomedical fields (19). The color of the obtained modified rice powders of the four varieties was off-white while that of the color of the original raw rice powders was pure white.

3.3. Morphology of rice particles

Particle morphology and approximate size of the raw rice powders in comparison with the modified rice powder under SEM analysis is demonstrated in Figure 1A. The raw rice particles of both glutinous and non-glutinous varieties displayed an irregular polygonal shape with several obvious sharp edge surfaces. Our findings on the shape of rice powders are similar to those previously reported (20), but the size of the rice particles was not in agreement. This may be due to the difference in rice varieties and the method of preparation of rice powders. A layered organization was clearly observed in NSP and some parts of JM particles. Small pieces, possibly "broken particles" produced by the preparation process were observed attached to the surface of the large particles (21). Small particles were more commonly observed in the glutinous than in the non-glutinous rice. Among the four rice varieties, JM particles showed the smallest size granules of about 3-6 μm whereas the average size of the others was approximately 5-10 μm . In comparison with the raw rice, the chemical modification under the present conditions caused a significant change to the rice particles as seen in Figure 1B. The modified JM particles were slightly swollen and the surface edges were blunt. The particles size was slightly larger (5-10 μm) than the raw rice (3-6 μm). The change in SH particles after modification was similar to that of JM in terms of particle swelling and edge blunting. However, modified SH particles also exhibited a high degree of alteration in terms of particle clustering or merging. Interestingly, the modification produced noticeable changes in glutinous rice varieties. The modified rice particles of both NSP and NKK displayed prominent changes in shape and surface with extremely higher swelling and merging than the non-glutinous rice, in that some particles of the rice appeared to be completely fused and that individual particles could not be observed. The surface of the modified NSP and NKK rice appeared rough and wrinkled. Uneven structure with fine porosity was clearly observed. It was proposed that the alkaline environment during the carboxymethylation reaction accounts for the structural changes. A previous report indicated that amylopectin is responsible for the swelling

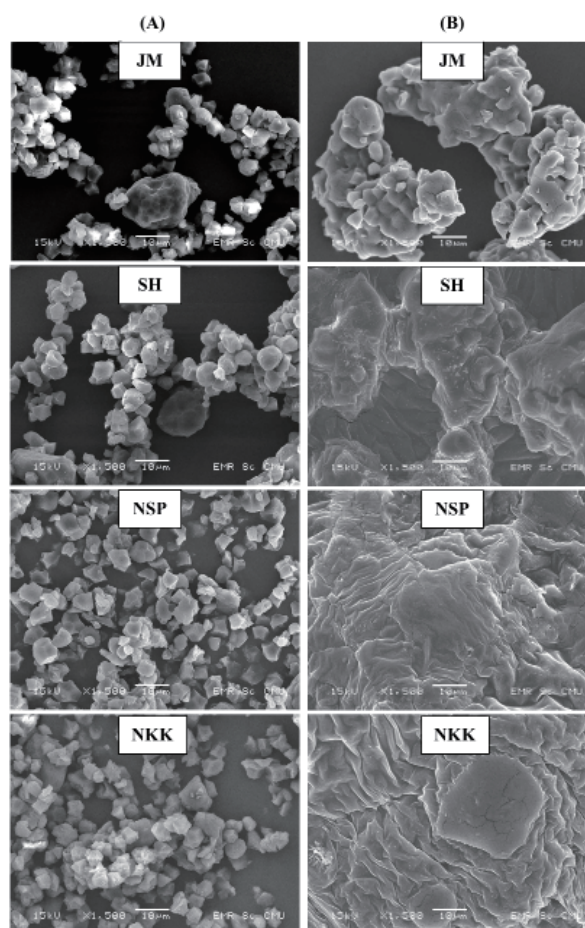


Figure 1. SEM micrographs of the raw (A) and modified (B) rice powders.

property of starch, whereas amylose acts as a connector to hold the starch particles intact (22). However, recent research reported that the extent of rice swelling after modification was due to the amylose content (20). In the present study, the glutinous rice which contained significantly less amylose demonstrated higher swelling and microstructural change than the higher amylose content, non-glutinous varieties like JM or SH. We propose that the results of the present study are due to the mutual activity of amylose and amylopectin content as well as the other components existing in the rice powders after modification.

3.4. Internal solid structure of rice particles

The XRD is used to reveal the presence and characteristics of internal crystalline structures of starch particles. Crystalline starch lattices are formed due to the arrangement of amylose and amylopectin molecules which can be classified to A-, B- and C-type by XRD pattern analysis (23). A-type starch has strong diffraction peaks at about 15° and 23° with unresolved doublet at around 17° and 18°. B-type starch gives a characteristic peak at about 6° and the strongest diffraction peak at

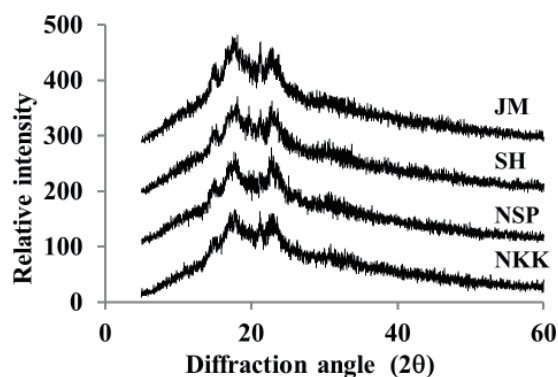


Figure 2. XRD patterns of the raw rice powders.

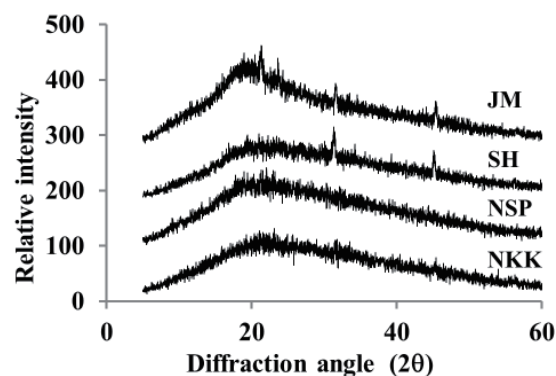


Figure 3. XRD patterns of the modified rice powders.

Table 2. XRD diffraction angles of the raw rice and solubility index of the rice powders

Rice variety	XRD diffraction angles (2θ)	Solubility index (%)	
		Raw rice	Modified rice
JM	14.9, 17.2, 20.5, 22.9	0.2 ± 0.1	97.6 ± 0.1
SH	14.8, 17.2, 20.5, 22.8	0.3 ± 0.1	99.8 ± 0.2
NSP	14.7, 17.1, 22.7	0.0 ± 0.0	93.1 ± 0.2
NKK	14.5, 16.9, 21.0, 22.7	0.0 ± 0.0	94.9 ± 0.1

around 17° with some small peaks at about 15°, 20°, 22°, and 24°. B-type starch is more resistant to enzyme hydrolysis than A-type starch (24). C-type starch is a mixture of A and B-type polymorphs and shows strong diffraction peaks at about 6° and 15° (according to B-type) and 17° and 23° (according to A-type). The C-type can be further classified as CA-type (closer to A-type) and CB-type (closer to B-type) according to the proportion of A- and B-type polymorphs. The XRD patterns of the raw rice samples of the four tested varieties are shown in Figure 2. It was found that all raw rice samples displayed strong reflections with different diffraction angles (Table 2). The diffraction peaks around 14.5-14.9°, 16.9-17.2° and 22.7-22.9° were considered to be the A-type crystalline arrangement. The minor crystalline peaks similar to B-type was observed in the XRD patterns of JM, SH and NKK around 20.5-21.0°. Starches from cereals usually present A-type structure. However, only NSP showed the characteristics of A-type structure. The crystalline structure of JM, SH and NKK were considered as CB-type because they showed one peak that seemed to be a typical of B-type. The different results of JM, SH and NKK from the previous reports on the type of crystallinity may have been largely due to differences in amylose content in the tested rice varieties, and to a lesser extent an effect of the process of rice powder preparation prior to XRD measurement.

After modification, the internal solid structure of the rice obviously changed. XRD halo patterns were found indicating the change of crystalline structure to an amorphous form. However, the level of crystalline destruction was different among the four rice varieties. As seen in Figure 3, the internal structure of the two

glutinous rice varieties were completely changed to an amorphous structure whereas some crystalline peaks were still observed in the non-glutinous varieties. All the original XRD crystalline peaks of SH disappeared after modification, and were replaced by new crystalline peaks at the diffraction angles of 31.2° and 45.0°. The XRD of the modified JM was similar to that of the modified SH as far as the new crystalline peaks, but one small original crystalline peak at 20.5° still remained. The XRD results were in agreement with the morphological changes observed in the SEM. The results suggested that the carboxymethylation conditions used in the present study caused the complete loss of crystallinity in glutinous rice varieties but not totally for the non-glutinous rice. It appears that the crystalline solid structure of the low amylose rice powders is easily destroyed by chemical modification. Some small peaks remaining in the XRD patterns of the modified rice of the high amylose varieties suggest the less destruction of amylose double helices, and that the new crystalline structure might be formed after chemical modification. These results suggest that the non-glutinous varieties need higher different conditions for chemical modification than the glutinous varieties.

3.5. Solubility index

Water miscibility is important for gelling substances in order to form desirable hydrophilic gels. Gelling agents with high water solubility can provide hydrophilic gels with the preferable characteristic of high transparency. In the present study, rice powders of the four varieties demonstrated significantly different solubility properties as shown in Table 2. It was also clearly seen that the modified rice powders have extremely higher aqueous solubility than the raw rice. This result confirmed that substitution of -OH with -CH₂-COO- group during rice modification enhances its water solubility. In addition, it is known that the granules disruption during modification highly increased the hydrophilic character of the starch. This effect also enhances the water solubility of the modified starches. The results demonstrated that the

non-glutinous varieties had significantly higher water solubility than the glutinous rice. It is also noted that between the two non-glutinous rice varieties, SH was higher soluble than JM. With respect to the glutinous varieties, NKK, whose amylose content is higher, showed higher water solubility than NSP. These results indicate that the solubility of modified rice powders is directly related to amylose level. These findings were in good agreement with those previously reported (25).

3.6. Appearance and internal solid structure of the gels

It is known that rice gels prepared by thermal pregelatinization gradually retrograde during storage. To retard or inhibit the retrogradation of rice starch molecules, chemical modification has been introduced to starch granules (26). In the present study, the four rice

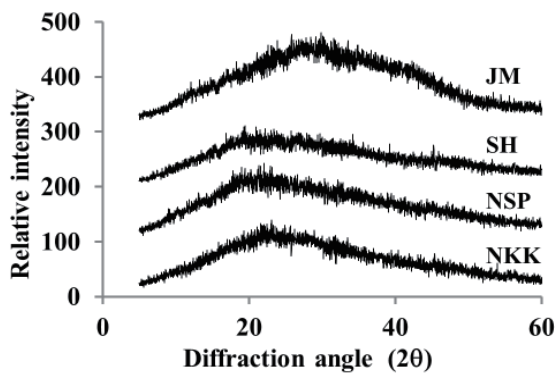


Figure 4. XRD patterns of the modified rice gels.

powders were chemically modified by etherification prior to gel preparation. All of the prepared gel formulations were similar in outward appearance as transparent semisolids and had a homogeneously desirable external structure. The non-glutinous gels were colorless whereas some color was observed in the glutinous NSP and NKK rice gels. The color of NSP gels was pale yellow while that of NKK gels was pale fulvous. The result of the internal crystalline structure analysis of the gels by XRD is presented in Figure 4. The XRD diffraction of all rice gels exhibited the complete halo patterns indicating that the crystalline structure of all modified rice particles was completely destroyed. It is noted that the crystalline structure of the modified non-glutinous powders disappeared when the rice powders were transformed into a gel structure. This result indicates that the condition of gel preparation as well as the concentration of rice powders in the gels was suitable and that the water quantity was sufficient for complete hydration of the rice particles to form the non-crystalline network structure of the gels.

3.7. Rheological behavior of the gels

Rheological property is very important for pharmaceutical gel formulations because this knowledge can provide the microstructural environment or flow behavior information of the gel which is responsible for drug diffusion and compatibility. The rheological behavior of the obtained rice gels is shown in Figure 5. The stress-strain relationship was not linear for all

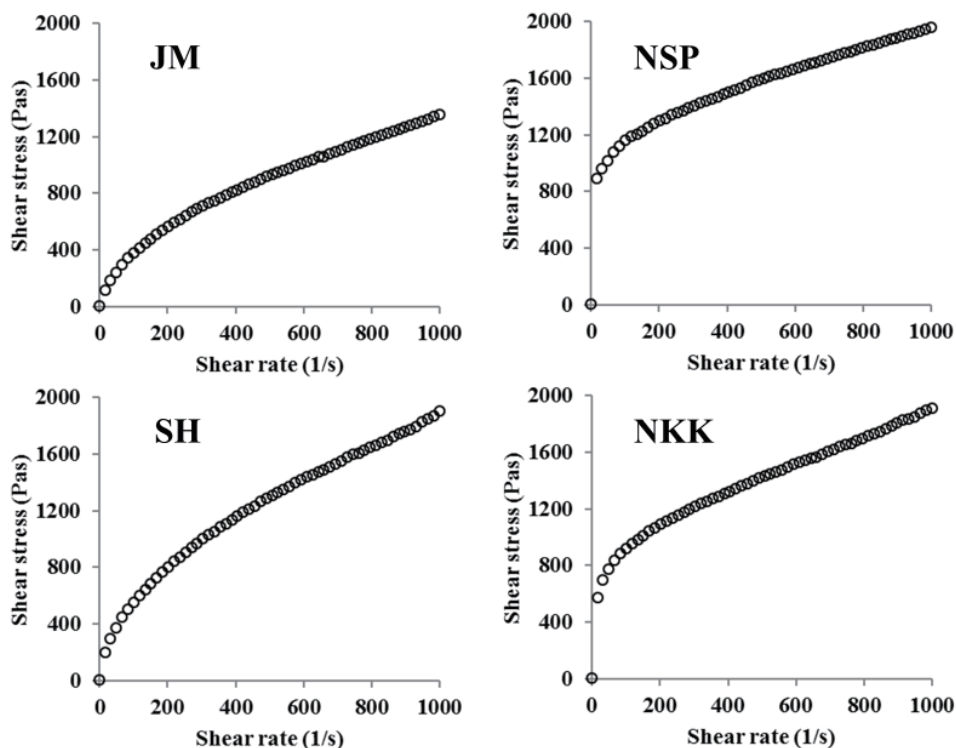


Figure 5. Rheograms of the modified rice gels.

Table 3. Rheological parameters and mucoadhesive properties of the modified rice gels

Rice variety	Rheological parameters		Mucoadhesion
	Viscosity (mPas)	Yield value (mPas)	Detachment time (min)
JM	2192.9 ± 54.7	→ 0	45 ± 2
SH	3167.4 ± 37.4	→ 0	56 ± 4
NSP	6213.1 ± 540.9	1077.4 ± 185.9	30 ± 3
NKK	4392.9 ± 315.3	536.1 ± 45.8	35 ± 2

samples suggesting that the rheological property of the obtained rice gels was a non-Newtonian flow and the gel viscosity was changed when the gels were sheared. This result indicates the incomplete formation of gel structure because of the lower shear stress. The non-glutinous JM and SH gels showed pseudoplastic flow indicating an immediate flow after stress application whereas the glutinous NKK and NSP gels showed plastic flow with yield value. Yield value is defined as the minimum stress that must be applied before the material really starts to flow (27). In pharmaceutical fields, it is very important to know the viscosity and yield value of the gel formulations because many passages (tubes, buccal applicators) are required for their packaging and administration. The viscosity and the yield value of the formulated gels are shown in Table 3. The viscosity of the glutinous rice gels was found to be significantly higher than that of the non-glutinous gels. Among the four varieties, the gel of NSP presented the highest viscosity of about 6 pascals-second (Pas) whereas that of JM displayed the lowest viscosity of approximately 2 Pas. Gels with yield value present advantage that the gels that do not flow out of their container due to their own weight if the container is inverted. However, the yield value of the developed glutinous gels is too high so as to offer significant resistance during application on the applied surfaces. Between the two glutinous varieties, the yield value of the NSP gel was significantly higher than that of NKK gel. Previous research reported that the rheological properties of the rice gels developed by thermal gelatinization were influenced by amylose content and concentration of starch (28). In the present study, the rice concentration in the gel formulations was fixed at 10%. Therefore, the variation of rheological behavior of the rice gels was considered to be influenced by mutual interaction of rice components particularly amylose content and water in the gels.

3.8. Mucoadhesive property of the gels

Mucoadhesive property is important in pharmaceutical formulations for trans-mucosal application in both localized and systemic drug delivery system. It was reported that the enhancement of drug delivery through mucosal membranes could be successfully achieved by ensuring the sufficient mucoadhesive strength of the delivery systems (29). Gels with high mucoadhesive

properties can be retained in the application area for the desired duration of action whereas those without this property are easily washed away and removed by the surrounding medium such as saliva, nasal or vaginal fluids in buccal, nasal, and vaginal cavities, respectively. Therefore, mucoadhesive property is an important issue for prepared gels administered *via* mucosal membranes. Table 2 illustrates the time required for gel detachment from the mucosal membrane after immersed in the water at body temperature with a stirring speed of 150 rpm. As can be seen, the non-glutinous rice gels showed significantly longer adhesion times indicating higher mucoadhesive strength than the glutinous rice gels. Moreover, it was observed that the non-glutinous rice gels were gradually dissolved in the medium until exhausted while the glutinous rice gels were ruptured by the stirring force and some small pieces were detached from the gels. Mucoadhesion is the interfacial force between the drug delivery system and the mucus layer coating an epithelium. Basic theories such as adsorption, wetting and diffusion phenomena have been described and associated with the mechanisms by which mucoadhesion occurs (30,31). These phenomena are enhanced by substances having more hydrophilic properties. As higher lipid content was found in the glutinous than in the non-glutinous varieties, it is thus considered to be a cause of lower adsorption force and wetting property as well as surface energy interactions of the glutinous gels when spreading onto the membrane.

4. Conclusion

The effects of rice varieties on the physicochemical characteristics of rice powder and properties of the derived gels obtained from the respective modified rice powders were explored in this study. The glutinous NSP and NKK and the non-glutinous JM and SH rice varieties yielded carboxymethylated modified rice powders with different morphology, crystallinity and aqueous solubility characteristics. Higher amylose content in the non-glutinous rice varieties significantly affected the internal crystalline structure of the rice powders and mucoadhesive as well as rheological properties of the respective derived gels. Rheological behavior of the non-glutinous JM and SH gels was pseudoplastic flow without yield stress whereas the glutinous NSP and NKK gels were more viscous and exhibited plastic flow with obvious yield values. These different gel properties reflect the different rice varieties used for gel preparation. The results suggest that non-glutinous rice varieties with high amylose contents are the most suitable for preparing pharmaceutical gels for trans-mucosal systems.

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