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Surface modification via alginate-based edible coating for enhanced osmotic dehydration mass transfer of ginger slices

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Abstract

Ginger has a high moisture content, which makes it highly susceptible to spoilage. Therefore, the shelf life can be extended through drying. In the drying process, osmotic dehydration is applied as pre-treatment due to its simple operation and energy-saving process for removing moisture from food. However, large solute gain during the osmotic dehydration has become the major challenge of this process as it has a negative impact on the final product. The edible coating is the key step to circumventing this issue. Alginate is a potential candidate for the coating material to enhance the mass transfer kinetics of the osmotic dehydration process. This study investigated the surface modification of ginger slices caused by the cross-linker calcium chloride and plasticizer glycerol on alginate coating using a Scanning Electron Microscope. Furthermore, the kinetics of water loss and solute gain were evaluated and modelling aspects were conducted. It was observed that the surface roughness of ginger coated with a combination of alginate, glycerol and calcium ions has reduced. This facilitated the mass transfer process, which was observed to have a high water loss and a lower solute gain. The Peleg model presented the best fitting model of mass transfer kinetics during osmotic dehydration of ginger slices. From this work, it can be deduced that alginate-based coating can be a promising pre-treatment step in the osmotic dehydration process.

Keywords

Ginger, alginate, coating, osmotic dehydration, mass transfer

Introduction

Ginger (*Zingiber officinale*) a member of the Zingiberaceae family, is widely used as a spice in foods and beverages throughout the world. It is also a good source of numerous bioactive phenolics, including non-volatile pungent compounds such as gingerols, paradols, shogaols and zingerones (1). In 2019, the total world production of ginger reached 4.08 million tonnes, with the majority of the production coming from India (2). Fresh ginger is reported to have a high-water content which is around 90% (3-5). Water provides conditions for microorganisms to grow and multiply, which causes the deterioration of food products and thus contributes to food wastage. Therefore, it is a vital step to lower the moisture content with appropriate preservation technologies.

Currently, a plethora of improved drying techniques has emerged in food processing to enhance the quality of food products and the sustainability of processing. Osmotic dehydration has received great attention as a pre-treatment step in the drying process due to its simple operation, where a partial amount of water is removed from a food product by immersion in a hypertonic solution. Moreover, it is also described as a quality improvement and an energy-saving process. Previously, it was reported that applying osmotic pre-treatment prior to microwave-assisted vacuum drying of blackberries results in reduced drying time and increased anthocyanin retention (6). In a study conducted, it was shown that osmotic dehydration improved the colour of apricots after hot air drying (7). A techno-economic study reports that by combining osmotic dehydration as pre-treatment prior to the drying process of broccoli stalk slices, the energy input can be reduced by 68% (8).

During the immersion time in the osmotic process, the two major mass transfers that occur simultaneously are the water outflow from the product to the solution and the solute inflow from solution to product. The former is known as water loss (WL) and the latter as solute gain (SG). The major challenge of the implementation of this process in the food industry was due to the large solute gain, which has a negative impact on the final product. To address this issue, edible coating is a key step to minimise solute gain during the osmotic dehydration process.

The edible coating is a thin layer applied to the product's surface to prevent the migration of moisture, oxygen and solutes into the food (9). Over the last decade, the production of edible coatings from food-grade biopolymers, in particular polysaccharide, has progressed significantly owing to increased interest in the food, pharmaceutical and cosmetics industries (10-12). Alginate is a natural polysaccharide extracted from brown algae that consists of a linear block co-polymer made of 1-4 linked β-D-mannuronic acid (M) and α-L guluronic acid (G) and widely used as a coating material due to its good film-forming ability (13). Formerly, the coating material was applied as a single-component formulation, but recent research has concentrated on developing the formulation of composite or multi-component edible materials with improved functional properties (14, 15).

As a natural biopolymer, alginate films are water-soluble, but they can be rendered insoluble by crosslinking with divalent cations or polyvalent cations such as calcium ions (16). When alginate crosslinks with calcium, a strong gel is formed and this could potentially improve the barrier and mechanical properties of alginate films. It has been reported that calcium chloride has the fastest gelation rate when compared to the other calcium used (17). Meanwhile, plasticizers are small molecules with low molecular weight and volatility that contribute to the strength and flexibility of polymeric materials (18). The incorporation of the plasticizer with the polymer matrix in terms of solubility and compatibility is important for effective plasticization (19). Polyols, organic esters and oils and glycerides are the 3 types of plasticizers that are commonly used. Glycerol, the water-soluble polyol, was found to be an excellent plasticizer for alginate films (20). Previously, an attempt was made to use glycerol as an osmotic agent to improve the performance of mass transfer during the osmotic dehydration process (21, 22). So far, the use of glycerol in coating formulations for accessing the mass transfer exchange during the osmotic dehydration process is scarce in the literature.

Therefore, the goal of this study was to investigate the effects of calcium as a cross-linker and glycerol as a plasticizer of alginate as an edible coating on the surface modification of ginger and its impact on the mass transfer kinetics of osmotic dehydration. In addition, the best mathematical model that can describe the mass transfer kinetics was also evaluated.

Materials and Methods

Materials

Sodium alginate (SA, CAS No. 9005-38-3), anhydrous calcium chloride ($CaCl_2$, purity ≥ 97%) and glycerol (ACS reagent, purity ≥ 99.5%) were obtained from Sigma Aldrich (Germany). Commercial sucrose (Prai, Malaysia) was used as the osmotic agent, and distilled water was used to prepare the coating and osmotic solution. Ginger was purchased from the local market in Kuala Nerus, Terengganu, Malaysia. The ginger was cleaned before being mechanically sliced to a uniform thickness of 1 mm and a diameter of 4 mm. Fresh ginger had a moisture content of 90.55 ± 1.55 % (wet basis), as determined by the Association of Official Analytical Chemists (AOAC) method (23).

Preparation of coating solution and coating process

The coating solution was prepared as per standard procedure (24). The 2% (w/w) of alginate solution was obtained by dissolving the alginate powder in distilled water and stirring continuously using a magnetic stirrer (WiseStir, MSH-200, Finland) at a temperature of 70 °C until a clear solution was achieved. The 2% (w/w) of $CaCl_2$ solution was prepared by dissolving the $CaCl_2$ powder in distilled water and stirring at room temperature. The solution of alginate and glycerol was prepared by mixing 2% of alginate and 2% of glycerol and dissolving the mixture with distilled water. The mixture was then stirred continuously at 70 °C until a clear solution was obtained.

Ginger slices were divided into 4 portions for coating process by dipping. The first portion was coated with only alginate. The second portion was initially dipped in alginate solution and then in $CaCl_2$ solution. The third portion involved dipping the ginger slices in a mixture of alginate and glycerol solution and then in a $CaCl_2$ solution. The last portion is the uncoated condition, which is used as a control. The dipping process for each solution took about 2 min and the excessive coating solution was drained off using a mesh tray before being dried in an oven (Memmert, UNB 100, Germany) for 10 min at a temperature of 55 °C.

Scanning Electron Microscope (SEM)

The uncoated and coated ginger was immersed in a 2.5% glutaraldehyde fixation in a 0.1 M sodium phosphate solu-

tion for 2 hrs at room temperature. All samples were rinsed 3 times with 0.1M sodium cacodylate buffer, pH 7.2 for 10 min. After that, the samples were post-fixed in phosphate buffer with 1% osmium tetroxide in the absence of light at room temperature and followed by an ethanol series dehydration (30, 50, 60, 70, 80, 90, 95 and 100%, keeping the sample for 15 min at 100%). Then, the samples were subjected to chemical drying via hexamethyldisilazane (HMDS) for overnight according to the standard procedure (25). Subsequently, each of the samples was coated with a layer of gold to produce electrical conductivity on the sample surface before being mounted onto an aluminium stub using double-sided sticky carbon tabs. The sample was scanned using a SEM (JEOL, JSM 6360 LA) operating at 10 kV in a high vacuum mode.

Osmotic dehydration

The 50% (w/w) osmotic solution was prepared by dissolving the sucrose in distilled water. The osmotic dehydration process was conducted by immersing the samples into the sucrose solution for 30, 60, 90, 120 and 150 min at room temperature. The ratio of sample to solution was set at 1:10. The sample was then separated from the solution, washed and rinsed with distilled water until the coating layer was removed. The surface was then wiped with tissue paper.

Mass transfer determination

The mass exchange between ginger slices and sucrose solution during osmotic dehydration was determined using the following equations (26):

$$Solid\ Gain\ (SG) = \frac{(W_{st} - W_{so})}{W_{wo} + W_{so}} \times 100 \quad \text{(Eqn. 1)}$$

$$Water\ Loss\ (WL) = \frac{(W_{wo} - (W_t - W_{st}))}{W_{wo} + W_{so}} \times 100 \quad \text{(Eqn. 2)}$$

where W_{wo} is the mass of water in sample before dehydration (g), W_t is the mass of the sample after dehydration (g), W_{so} is the mass of the solids in the sample before dehydration (g) and W_{st} is the mass of the solids in the sample after dehydration (g).

Mass transfer kinetics modelling

The empirical models for osmotic dehydration listed in Eqn. 3 to 5 were used to predict the kinetics of water loss and solute gain during the osmotic dehydration process of ginger slices (27).

$$Peleg\ model = \frac{t}{k_1 + k_2 t} \quad \text{(Eqn. 3)}$$

$$Magee\ model = A + k\sqrt{t} \quad \text{(Eqn. 4)}$$

$$Penetration\ model = k\sqrt{t} \quad \text{(Eqn. 5)}$$

Where A and k are constant and t is time (min).

The statistical parameters such as determination coefficient (R^2), reduced chi-square (X^2) and root mean square error (RMSE). The R^2 at its highest and the lowest X^2 and RMSE values required for assessing fitness were used.

$$R^2 = 1 - \frac{\sum_{i-1}^{i=n}(X_i^{exp} - X_i^{pred})^2}{\sum_{i=n}^{i=n}(X_i^{exp} - \bar{X}_i^{pred})^2} \quad \text{(Eqn. 6)}$$

$$\chi^2 = \sum_{i-1}^{i=n} \frac{(X_i^{pred} - X_i^{exp})^2}{n - m} \quad \text{(Eqn. 7)}$$

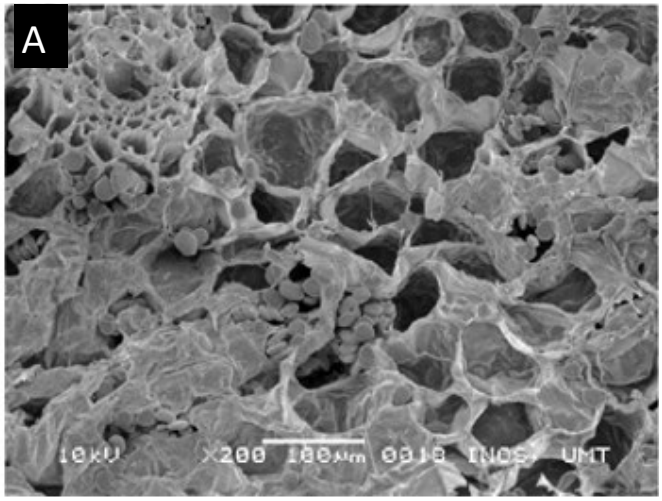
$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(X_i^{pred} - X_i^{exp})^2} \quad \text{(Eqn. 8)}$$

where n is the number of observations, $X^{pred}{}_i$ is the WL or SG predicted i^{th} value, is the WL or SG, $X^{exp}{}_i$ experimental i^{th} value, m is the number of variables and X^{exp} is the mean of the WL or SG experimental value.

Results and Discussion

Microstructure analysis (SEM)

The SEM micrographs demonstrated the microstructure of uncoated ginger and different alginate coating conditions of ginger slices are presented in Fig. 1. The Fig. 1 (A) shows the surface structure of uncoated ginger slices where cellulosic walls and starch granules can be clearly seen in the micrographs. As alginate was applied as a coating material, a gel structure was observed on the surface of the ginger, as shown in Fig. 1 (B). Due to weak coating-sample adhesion, the surface appears to be rough and non-smooth. According to previous research, the presence of a hydrophilic cut surface on the sample makes adhesion extremely difficult due to the low surface energy (14, 28). As shown in Fig. 1 (C), the presence of calcium as a divalent cation induces gelation of alginate and the surface roughness increases. It was found



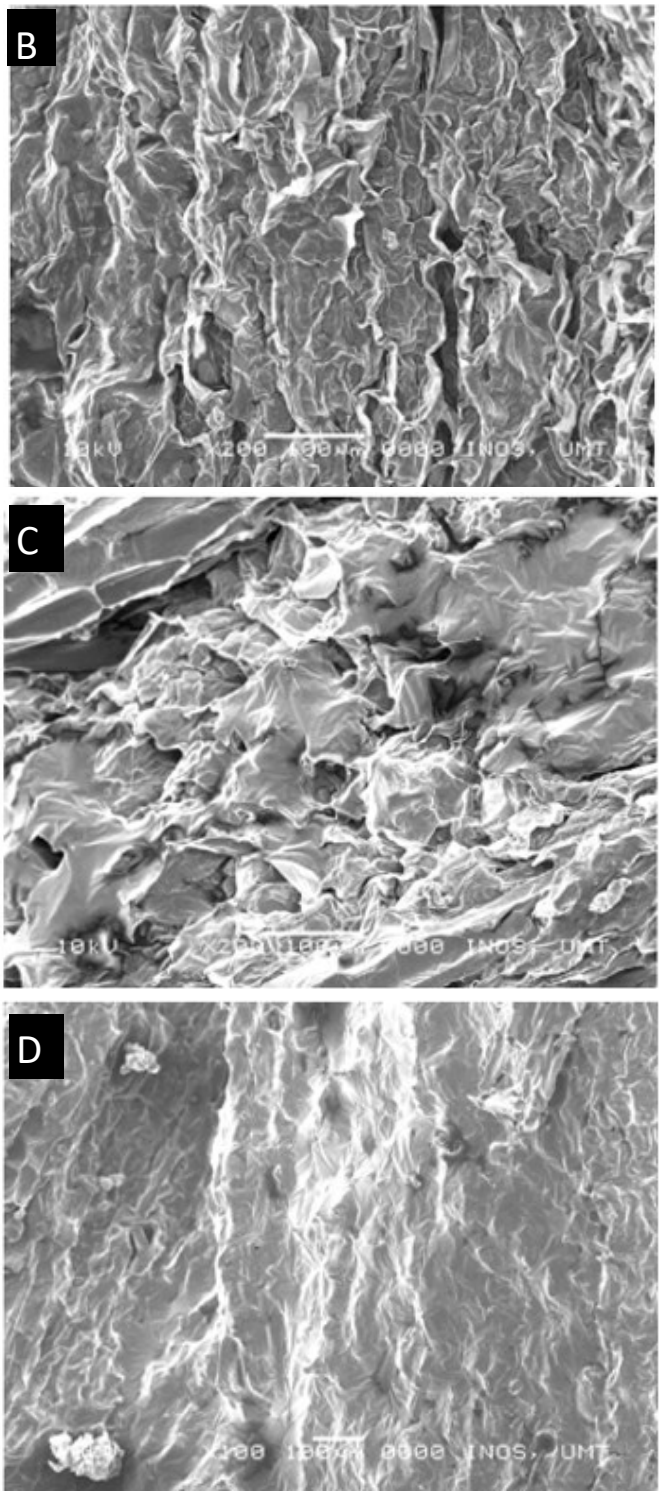


Fig. 1. The microstructure of surface morphology of (**A**) Uncoated, (**B**) Alginate coated, (**C**) Alginate + $CaCl_2$ coated and (**D**) Alginate + glycerol + $CaCl_2$ coated.

that calcium increases crosslinking in the polymer chain, which makes the intermolecular bonds more cohesive (29). Meanwhile, good adherence of the surface coating was seen on a combination of alginate, glycerol and $CaCl_2$. The addition of glycerol to the coating formulation results in more pronounced changes to the surface coating, as illustrated in Fig. 1 (D) where the surface roughness is reduced. The small molecular chains in glycerol allow it to penetrate polymer networks, occupying space more easily through hydrogen bonds and causing the polymer structure to become flexible (30).

Water loss (WL)

The kinetics of WL during osmotic dehydration of ginger slices are shown in Fig. 2. The gradual increase in WL can be observed for all process conditions when immersion time is increased during the OD process. Rapid water removal was reported on various agricultural products for the first 2 hrs of processing time due to a high osmotic driving force between the sample and the hypertonic solution (31-33). From the graph, it is apparent that the WL increased for all coating conditions when compared to the uncoated sample. This indicates that the hydrophilic nature of alginate gels enables the transfer of water molecules (34). This finding is consistent with an earlier study (35) which was also found that higher WL was obtained when an alginate coating was applied during the osmotic dehydration process of pumpkin cubes.

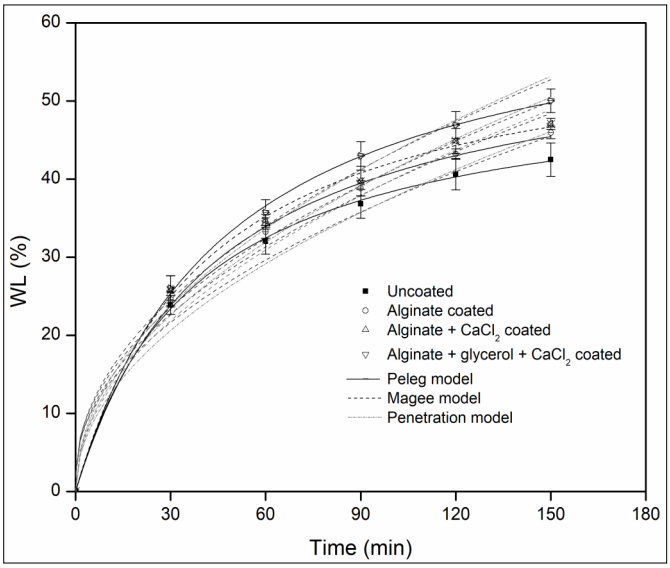


Fig. 2. The kinetics of water loss (WL) during osmotic dehydration of ginger slices.

On the other hand, the surface modification caused by the addition of $CaCl_2$ as a cross-linker and glycerol as a plasticizer had a correlation with the WL performance. The calcium-induced gelation of alginate was sufficiently strong to maintain the osmotic pressure of the sucrose solution, resulting in a higher WL than alginate-coated. It was reported that the "egg structure" formed by calcium ions and alginate solution allows water to pass through the coating with significantly greater ease during the osmotic dehydration process of strawberries (36).

Surprisingly, the proportion of glycerol in the coating formulation of ginger samples has a higher WL value of 50%. It has been confirmed in this study that the presence of glycerol in the polymeric structure results in improved adhesion and greater flexibility of the coating, which helps to maintain osmotic pressure and thus improves the performance of WL. Glycerol is also hydrophilic in nature, giving the polymer molecules greater affinity to bind water within the structure (37, 38).

Solute gain (SG)

The kinetics of SG during osmotic dehydration of ginger slices is depicted in Fig. 3. During the first 30 min of immersion time, non-coated samples presented significantly in-

creased SG values compared to coated samples. After 150 min of immersion time, the SG values were around 6.3% for the uncoated sample. These findings indicate that a significant amount of SG can be minimised once the coating is applied to the ginger slices. The layer of coating and the accumulation of solids on the coating surface may act as hindrances to SG during the osmotic process. These results match those observed in earlier studies on strawberries (36).

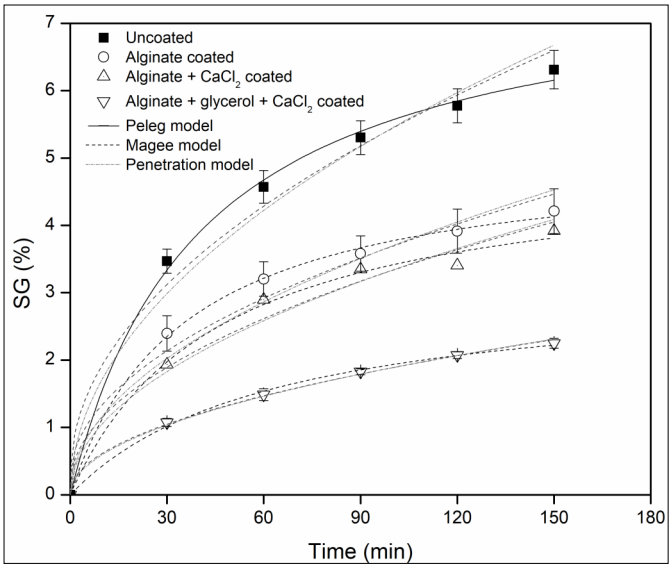


Fig. 3. The kinetics of solute gain (SG) during osmotic dehydration of ginger slices.

In the case of coated samples, SG was slightly higher in the absence of glycerol. This result shows that the combination of alginate, glycerol, and $CaCl_2$ as coating material results in an improved mass transfer barrier with only 2% of SG. Remarkably, the SG obtained in this study was also significantly lower than the SG obtained in a previous study that used carboxymethyl cellulose as a coating material for osmotic dehydration on ginger slices (39). As a result of these findings, it is clear that the component's effect on the surface tension of the coating solutions is crucial. Thus, better uniform spreading ability of the edible coating has become a desired property when creating edible coating formulations.

Modelling of osmotic dehydration mass transfer kinetics

Mathematical modelling is required to determine whether a model adequately describes a process and to estimate model parameters accurately. Fig. 2 and Fig. 3 depict the curve fitting of the three models that were considered in this study for WL and SG respectively. Curve fitting statistics for the Peleg models are presented in Table 1. The Peleg model constant is related to the mass transfer rate, where k_1 is inversely related to the initial mass transfer rate and the k_2 values inversely describe the values at equilibrium. For WL, the k_1 did not exhibit a trend when conditions were compared. However, the value of k_2 was decreased accordingly as coating material was proportionately applied, indicating that a high amount of water was removed from the products. In contrast to SG, both k_1 and k_2 increased when coating was applied, noting that the lower solute was diffused into the products.

The parameters obtained for WL and SG using the Magee model are presented in Table 2. For parameter *A*, which represents the contribution of hydrodynamic mechanism due to capillary pressure at a very short time did not demonstrate a trend for both WL and SG. However, the rate of mass transfer rate which represent as parameter *k*, showed an increasing trend for WL and decreasing trend for SG as coating material was proportionately applied to ginger slices. The penetration model parameters obtained from the curve fitting statistics are shown in Table 3. For WL, the *k* value increased as the coating material was applied. A higher *k* value can be seen under alginate + glycerol + $CaCl_2$ coated conditions for WL. While for SG, the opposite trend was observed.

For all conditions, the Peleg model exhibits an excellent fit of WL and SG experimental data compared to the Magee and Penetration models. The R^2 value of the Peleg fits was determined to be the highest at 0.99, while X^2 and RMSE were the lowest under all conditions. The X^2 and RMSE for Magee and the Penetration models are slightly larger than the Peleg model. As previously reported, the Peleg model has proven to be an excellent model for predicting the mass transfer of the osmotic dehydration process (40-42).

Conclusion

The alginate-based coating shows a promising step in improving the osmotic dehydration performance. Alginate as a single component coating enhances the mass transfer when compared to an uncoated sample. The addition of $CaCl_2$ to strengthen the alginate gel positively improves the WL and SG performance. Ultimately, the addition of glycerol to the alginate solution and its combination with $CaCl_2$ changed the surface of the ginger slice, resulting in a higher WL and a lower SG compared to the other conditions. Among the mathematical models used in this study, the Peleg model adequately described the mass transfer kinetics of WL and SG during osmotic dehydration of ginger slices under all conditions.

Table 1. Value of Peleg model parameters for water loss (WL) and solute gain (SG).

Condition	WL (%)					SG (%)				
	k_1	k_2	X^2	RMSE	R^2	k_1	k_2	X^2	RMSE	R^2
Uncoated	0.7166	0.0188	0.2095	0.4577	0.9991	5.1432	0.1281	0.0165	0.1284	0.9975
Alginate coated	0.7492	0.0170	0.4245	0.6516	0.9988	6.8805	0.1963	0.0042	0.0650	0.9985
Alginate + $CaCl_2$ coated	0.6995	0.0167	0.6161	0.7849	0.9983	9.1690	0.2010	0.0148	0.1217	0.9942
Alginate + glycerol + $CaCl_2$ coated	0.7241	0.0152	0.3011	0.5487	0.9993	20.0163	0.3167	0.0018	0.0428	0.9978

Table 2. Value of Magee model parameters for water loss (WL) and solute gain (SG).

Condition	WL (%)					SG (%)				
	A	***k***	χ^2	**RMSE**	R^2	***A***	***k***	χ^2	**RMSE**	R^2
Uncoated	2.3094	3.5268	6.5625	2.5617	0.9790	0.3071	0.5137	0.1047	0.3235	0.9841
Alginate coated	1.9136	3.7959	4.4749	2.1154	0.9875	0.2525	0.3439	0.0733	0.2708	0.9754
Alginate + $CaCl_2$ coated	2.1753	3.8983	5.8120	2.4108	0.9809	0.1439	0.3190	0.0516	0.2272	0.9798
Alginate + glycerol + $CaCl_2$ coated	1.7804	4.1594	4.6758	2.1623	0.9864	0.0297	0.1858	0.0015	0.0396	0.9981

Table 3. Value of Penetration model parameters for water loss (WL) and solute gain (SG).

Condition	WL (%)				SG (%)			
	k	χ^2	**RMSE**	R^2	***k***	χ^2	**RMSE**	R^2
Uncoated	3.7624	6.6536	2.5794	0.9734	0.5450	0.1085	0.3295	0.9794
Alginate coated	3.9911	4.5436	2.1315	0.9842	0.3697	0.0754	0.2747	0.9684
Alginate + $CaCl_2$ coated	4.1203	5.8949	2.4279	0.9806	0.3337	0.0467	0.2162	0.9772
Alginate + glycerol + $CaCl_2$ coated	4.3410	4.5748	2.1389	0.9867	0.1888	0.0014	0.0386	0.9978

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Authors contributions

MHH, KHK, AR, MNL, MINMI and NSMS conceptualized, designed and coordinated the study. MHH conducted the experimental works. All authors contributed to writing the manuscript, reviewed and approved the final version of the manuscript.

Compliance with ethical standards

Conflict of interest: Authors do not have any conflict of interests to declare.

Ethical issues: None.

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