IOP Conference Series: Materials Science and Engineering

PAPER • OPEN ACCESS

Moisture content and mechanical properties reduction of hard capsules upon prolong drying process

To cite this article: Mohd Aiman Hamdan *et al* 2021 *IOP Conf. Ser.: Mater. Sci. Eng.* **1092** 012057

Moisture content and mechanical properties reduction of hard capsules upon prolong drying process

Mohd Aiman Hamdan[1], Nur Hikmah Sulaiman[2], Khairatun Najwa Mohd Amin[1], Fatmawati Adam[1,3*]

[1]Faculty of Chemical & Process Engineering Technology, Universiti Malaysia Pahang, Lebuhraya Tun Razak, 26300 Kuantan, Pahang, Malaysia
[2]Academy of English Studies, MARA University of Technology, Terengganu Branch, Dungun Campus, 23000 Dungun Terengganu
[3]Centre of Excellence for Advanced Research in Fluid Flow (CARIFF), Universiti Malaysia Pahang, Lebuhraya Tun Razak, 26300 Kuantan, Pahang, Malaysia

Corresponding author: fatmawati@ump.edu.my

Abstract. Drying is a traditional preservation method to control the properties and quality of pharmaceutical product such as hard capsule. In this study, carrageenan and nanocellulose (NC) solution were mixed with crosslinker and toughening agents in the formulation solution and dipped for hard capsule formation. The dipping bar was dried at different drying times from 40-60 minutes in an oven. The properties of carrageenan hard capsule were compared with commercial hard capsule materials which were gelatine and hydroxypropyl methylcellulose (HPMC). Prolong drying time decreased the moisture content and hydroxyl group absorbance of carrageenan hard capsule up to 20 % and 2%, respectively. The mechanical properties of the hard capsule sample reduced about 19.2 % due to reduction of intermolecular interaction between carrageenan, crosslinker and NC. Increment drying time leads to tissue shrinkage in carrageenan hard capsule thus collapse the biocomposite structure and mechanical properties. Gelatine and HPMC hard capsules had lower moisture content and higher mechanical strength and thermal properties as compared to carrageenan hard capsule. As the drying time increase, case hardening phenomena collapses the structure and change the structure chain in the biocomposite.

1. Introduction

The application of natural polymers such as carrageenan, gums, alginic and starch in food, pharmaceutical and biomedical area is limited due to their poor mechanical strength, thermal and structural stability [1]. Carrageenan has a large potential natural polymer to be developed as a packaging in a food and pharmaceutical products. This is due to its demands, availability, and low-cost raw materials. However, the carrageenan tends to aggregate and produce a wrinkle and break film because of its low mechanical strength and complex natural behavior [2]. Thus, efforts have been made in terms of polymeric blending by addition of filler, and chemical or physical crosslinking to overcome these drawbacks [1]. Physical crosslinking will improve the mechanical and chemical stability of carrageenan biocomposites. Incorporation of physical crosslinker will create an intermolecular interaction between

carrageenan, filler and crosslinker (Hamdan, Lakashmi, Mohd Amin, & Adam, 2020). Physical crosslinker will create a bridge through hydrogen bonding interaction between helices of carrageenan, while the filler will fill in the empty void space between carrageenan and crosslinker[4].

Natural filler from plant based is commonly used to improve the stability and strength of natural polymer. In addition, natural filler from fiber could play an important role in developing biodegradable packaging material. This is due to its availability, strength and cost, besides resolving the current ecological and environmental problems [5]. The filler from fiber is utilized to enhance the mechanical and physical properties of the composites and to make the final product more economical. The natural filler is expected to assist in retaining the water molecule in the biocomposite thus improve the stability of the developed product.

Cellulose is one of the natural fibers and is the most abundant biopolymer on Earth. Cellulose is the main component of plant cell walls and the basic building block for many textiles and paper. It is abundantly available of all naturally occurring organic compound [6]. Nanocellulose (NC) is the nano size of the cellulose that ranged from 5 to 70 nm in widths and between 100 nm and several micrometers in lengths [7]. NC is commonly used as a nanofiller in composites such as hydrogel, cements, polymers, and others. This is due to its potential advantages which include mechanical, functional, biocompatible and biodegradable properties [8].

Even though the improvement in terms of mechanical properties was a success as biocomposite, but the shelf life of hard capsule is crucial to ensure the product's stability and quality as the hard capsule will be consumed by the body. One of the important parameters to monitor the shelf life of hard capsule is by manipulation of drying time. Drying process means removal of water from a material to preserve and maintain its quality for a long-term. The water content reduction slows down the biological activity, as well as chemical, and physical reaction that causes deterioration in the product during storage [9]. The most common methods used for drying are natural drying (drying in the shade or sun drying) and hot-air drying [10]. Drying is not only affecting the water content of the product but also alters physical, biological and chemical properties, such as enzymatic activity, microbial spoilage, hardness, aroma, flavor and palatability of foods. The removal of moisture in food and pharmaceutical products will prevent the growth and reproduction of the microorganisms. It will cause product decay thus minimizes many of the moisture-mediated deteriorative reactions [11, 12]. However, uncontrolled drying time will lead to a low quality end product with unfavorable appearance such as low mechanical strength due to undesirable physiochemical changes [13]. In fact, the increment of drying time will decrease the moisture content and mechanical properties of the hard capsule due to a high stress in the cellular building and case hardening phenomena [14].

In this work, drying time of carrageenan hard capsule incorporated with NC filler was studied. The effect of manipulating the drying time on the quality and stability of the biocomposite hard capsule was analyzed. The chemical and mechanical properties were compared to the commercial hard capsule materials which were gelatin and hydroxypropyl methylcellulose (HPMC).

2. Materials and Method

2.1. Materials

Semi refined carrageenan was supplied by TACARA, Sabah, Malaysia. Food grade crosslinker, alginic acid, titanium dioxide (TiO2), and microcrystalline cellulose (MCC) were obtained from Sigma-Aldrich, USA, while the plasticizer (PEG400) was obtained from Merck, Germany. Nanocellulose (NC) was obtained via ultrasonication of MCC for 50 minutes at an amplitude of 20% in deionized water (Amin et al., 2016; Hamdan, Adam, & Mohd Amin, 2018).

2.2. Preparation of nanocellulose-carrageenan hard capsule

Semi refined carrageenan, crosslinker, nanocellulose (NC), PEG, and alginic acid were mixed with deionized water in a double jacketed glass reactor. The formulation solution was mechanically stirred at

60 °C for 3 ½ hours. The hard capsule solution was dipped using self-fabricated capsule dipping machine using size "1" capsule shell.

2.3. Carrageenan hard capsule drying

The pin bar was dried at a constant temperature, 60 °C for three different drying times- 40, 50, and 60 minutes in the drying oven (BINDER, Germany). The hard capsule formulation was labeled as DT40, DT50, and DT60. The hard capsule physicochemical and mechanical properties were compared with commercial hard capsule materials which were gelatin and HPMC.

2.4. Hard capsule characterization

The moisture content of the hard capsule was determined using a moisture analyzer (AND MS-70, Japan). Approximately 0.1-0.5 g of sample was placed on the heating pan of the moisture analyzer. The moisture from the sample evaporates because of continuous heating. The analysis stopped automatically once the mass of the sample attained a constant moisture content value.

2.5. Functional group determination

A Perkin Elmer ATR-FTIR spectrometer (Frontier, USA) was used to analyzes the functional group presence in the hard capsule sample. FTIR was operated at spectra ranges of 400 to 4000 cm-1. A total of 32 scans were acquired at 0.15 s/scan and with a spectral resolution of 8 cm-1. The spectra were analyzed using OMNIC software.

2.6. Mechanical strength analysis of hard capsule

Two types of mechanical strength analysis were conducted using CT3 Texture Analyzer (Brookfield, USA) which were capsule loop tensile test and breaking force test. The texture analyzer was loaded with a 50 kg load cell. An average of at least three measurements was taken for each formulation. Texture analyzer was fitted with TA-CLT separating rod fixture for capsule loop tensile test. Testing mode of 'tension' and target option of 'distance' were selected. Upon operation, the probe will travel upwards at speed of 0.5 mm/s until the capsule was pulled apart and broke [17, 18]. The applied force was recorded as capsule loop strength.

For breaking force test, a flat-end probe (TA-10) with a diameter of 12.7 mm was used. Hard capsule was placed at the center of the texture analyzer platform. Testing mode of 'compression' and target option of 'distance' approximately 4.0 mm was programmed. Upon analysis at the speed of 1.0 mm/s, the probe was compressed onto the capsule and the breaking force was recorded [19].

2.7. Thermal analysis

Differential scanning calorimeter (DSC) was used to study the temperature and heat flow associated with the material transition of hard capsule sample. Approximately 5-10 mg sample was heated at 10 °C/min up to 300 °C using DSC (TA/Q 1000 DSC Series, Newcastle). The equipment was operated under heat/cool/heat mode.

2.8. Statistical analysis

The experimental works were conducted in triplicate and the data were presented as means ± standard deviations using OriginPro 9.0 software.

3. Results and Discussion

3.1. Moisture content analysis

A drying process involves a reduction in overall moisture content of a material. Besides, drying will keep or enhance the desired quality attributes in a cost-effective manner [20]. Determination of moisture content is important as the stability and shelf life of hard capsule depends on the amount of water contained in the product. Higher moisture content in food or pharmaceutical products is not good. This is because the products are susceptible to microorganisms which decrease their shelf life [21]. Critical moisture content is varied for different food or pharmaceutical products. Commonly, moisture content less than 15-16% is considered safe to prevent the growth of microorganisms that may lead to product contamination [22, 23]. Meanwhile, the moisture content of native cellulose is effectively removed at 60 ºC using tray drying method [24]. Table 1 represents the moisture content of carrageenan, gelatin and HPMC hard capsules. Increasing the drying time causes a decrease in the moisture content of the hard capsule. As the drying time increase from 40 to 60 minutes, the moisture content decreases from 17.51% to 13.95%. DT40 has higher moisture content as compared with GHC hard capsule. Meanwhile, the moisture content of HPMC hard capsule is lower than the DT50 and DT60 hard capsules. The result is in agree with a study conducted on jujube fruit, bread and bamboo shoot [12, 21, 25].

Table 1. Moisture content of carrageenan, gelatin and HPMC hard capsules

Formulation	Moisture Content (%)
DT40	17.51 ± 0.484
DT50	14.61 ± 0.561
DT60	13.95 ± 0.665
GHC	16.41 ± 0.911
HPMC	11.64 ± 1.050

Reduction of moisture content is probably due to the case hardening phenomena that occur on the surface of the hard capsule during the drying process [12]. Case hardening causes the loss of water from the surface layer to the surrounding. At a longer drying time, the product's moisture content is low due to the removal of water. Additionally, it is due to the maintenance of the porous structure during osmotic treatment [26]. The phenomena led to the development of locked-in pores caused by cellular collapse during dehydration [25].

Furthermore, at a low drying rate (low temperature), the moisture gradient within the product is small and internal stress is low [27]. Hence, the material will shrink down fully onto a solid core. Thereby, the water molecule in the porous media had moved to the surface layer of sample film followed by water evaporation [27]. After a critical point, the film surface will become stiff and limiting the shrinkage. Thus, the stress is higher resulting in increasing pore formation and affecting the film properties. In fact, the thickness of the product will also affect the resulting moisture content. As the thickness of the product increased, the time required to achieve a certain moisture content increased [28]. Besides that, moisture content found in a range of 36 to 38% may inhibit microbial growth thus prolong the shelf file of the products [21]. Based on the results in Table 1, shows that the developed carrageenan hard capsule is a stable product. Its low moisture content can prevent the carrageenan hard capsule from the microbial growth that may lead to product contamination.

3.2. Functional group determination

Fourier transforms infrared analysis was conducted to study the presence of specific functional groups in hard capsule biocomposite. Figure 1 shows the infrared spectroscopy of carrageenan hard capsule at different drying times, gelatin and HPMC hard capsules. The wavelength between 3500 to 3200 cm^{-1}, 1335 cm^{-1}, and 1160 cm^{-1} at DT40, DT50 and DT60 are associated with to the vibration range of intra- and intermolecular hydrogen bonded OH groups from the NC [29]. Meanwhile, the band absorption at

3430 cm^{-1} represents the amorphous properties of cellulose. It is attributed to the OH stretching related to the intra-molecular hydrogen bonds at the C3 position. It may also possible for intra-molecular hydrogen bonds between the functional groups at the C2 and C6 positions of the cellulose [29].

Presence of C-O of 3,6-anhydrogalactose and C-O-SO_4 on galactose-4-sulphate from carrageenan are observed at a wavelength of 922 cm^{-1} and 841 cm^{-1} respectively [30]. Meanwhile, the functional group for D-galactose-4-sulphate, 3,6 anhydro-D-galactose, glycosidic linkage and ester sulphate are attributed at a wavelength of 1033 cm^{-1} and 1220 cm^{-1}, respectively [31]. As the formulation of carrageenan and NC were kept constant throughout the study, no switch of wavelength was observed from the spectrum.

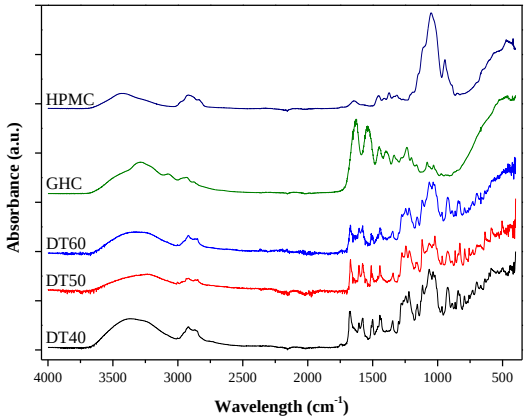

Figure 1. FTIR analysis of carrageenan hard capsule biocomposites, gelatine, and HPMC hard capsules

Absorbance area is determined to estimate the intensity and influence of network formation which is represented by each functional group [15, 18]. It can be analyzed by calculating the area under the curve at a specific range of wavelength as shown in Table 2. Increment of drying time from 40 to 60 minutes reduced the absorbance of hydroxyl group from 471.13 to 460.55 cm^{-1}. The result is in parallel to the decrease in the moisture content in the previous section. It proved that the manipulation of drying time affected the absorbance of hydroxyl group in the carrageenan hard capsule. This was due to the loss of water from the surface of hard capsule to the surrounding as the occurrence of evaporation phenomena in the system during the drying process.

Table 2. Absorbance area of carrageenan based hard capsule composites, gelatine and HPMC hard capsules

Sample	**Absorbance Area (cm^{-1})**		
	O-H	C-O	C-O-SO_4
DT40	471.13	49.22	52.98
DT50	470.45	42.97	45.26
DT60	460.55	41.93	43.43
GHC	403.23	40.08	No peak

HPMC	372.09	No peak	40.30

Absorbance areas of hydroxyl group in gelatin and HPMC hard capsules were lower. It represented that the product had low water content. This result proved that the commercial hard capsule products had a longer shelf life. Thus, it could prevent the growth of microorganisms that could cause contamination of the hard capsule. The spectroscopy showed that carrageenan hard capsule contains chemical groups that were also present in both gelatin and HPMC hard capsule. The wavelength at 840-850 cm^{-1} represents the C-O-SO_4 group and at 925-935 cm^{-1} represents the C-O group.

3.3. Mechanical analysis

In capsule loop strength test, texture analyzer will pull the capsule apart and the force required to cause rupture to the capsule was measured. Meanwhile, capsule hardness test was used in pharmaceutical industry to test the breaking point and structural integrity of the capsule before use [32]. Figure 2 represents the capsule loop strength and capsule hardness test for carrageenan based hard capsule at different drying time. The graph also represents the relationship between the moisture content and mechanical properties of biocomposites. As the moisture content increases, it produces a high mechanical strength of composite. The capsule loop test and capsule hardness test for GHC is 115.92 N and 17.11 N respectively. Meanwhile, the capsule loop test and capsule hardness test for HPMC is 72.59 N and 9.23 N respectively.

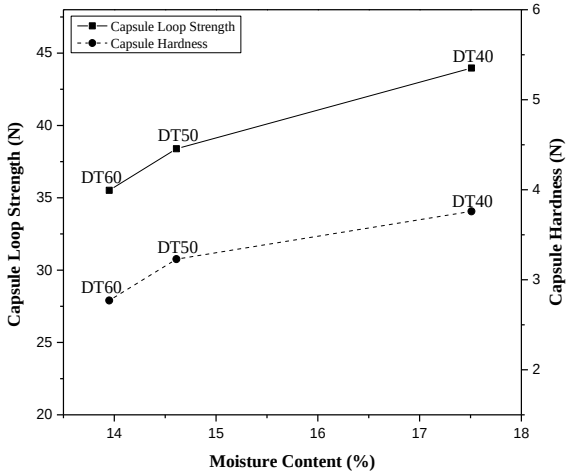

Figure 2. Capsule loop strength and capsule hardness test for carrageenan based biocomposite

The graph shows that, as the drying time increase from 40 to 60 minutes, the capsule loop strength of the hard capsule decrease from 43.9 N to 35.5 N. The presence of hydrogen bonding in the carrageenan biocomposite promotes the stability [33]. The reduction of mechanical properties may be attributed by the fragmentation of the carrageenan molecule that caused the degradation in the carbohydrate backbone [34]. Besides, this work suggests that upon prolong drying time, water molecules in the carrageenan hard capsule evaporate to surrounding. As a result, hydrogen bonding interaction between the carrageenan, crosslinker, and NC reduced, as proven by infra-red spectroscopy result. Due to low interaction between molecules, only a small amount of force is required to break the biocomposite hard capsule.

Hardness is defined as a resistance to an indentation in specimens [35]. The graph in Figure 2 shows that the hardness of hard capsule increase as the drying time increase. At 40 minutes, the capsule hardness was only 3.44 N. When the drying time increased to 60 minutes, the capsule hardness decreased up to 19% to 2.78 N. As the drying time increase, it creates a formation of a tighter structure and restricts the movement of molecules in the carrageenan network [29]. Thus, it leads to the reduction of moisture

content and case hardening phenomena occurrence, where more water molecules are lost from the hard capsule surface. As the water molecule reduced, the hydrogen bond interaction between molecules was also reduced and lead to the reduction of hard capsule hardness.

In addition, crosslinking of carrageenan with crosslinker in the matrix also affects this situation. Crosslinking prevents the swelling of carbohydrate and restricts the movement of water molecules in the matrix [29]. Thus, it led to a decrease in the amount of absorbed water and in agreement with the mechanical properties of the carrageenan biocomposites.

3.4. Thermal analysis

Upon drying process, several reactions are set off including chemical reactions, cellular collapse, and first and second order phase transitions [36]. As a result, structural property changes may occur in the composite.

Figure 3 represents the first heating of DSC thermogram for carrageenan based hard capsule at different drying times. A melting temperature, Tm in DSC thermogram is defined as the inflection point of the curves [37]. The result showed that as the drying time increased, the melting point of the biocomposites was decreasing from 199.5 to 190.4 °C. Increasing drying time caused severe tissue shrinkage in the composite, thus collapse the structure and lead to the change in structure chain in the biocomposite [25].

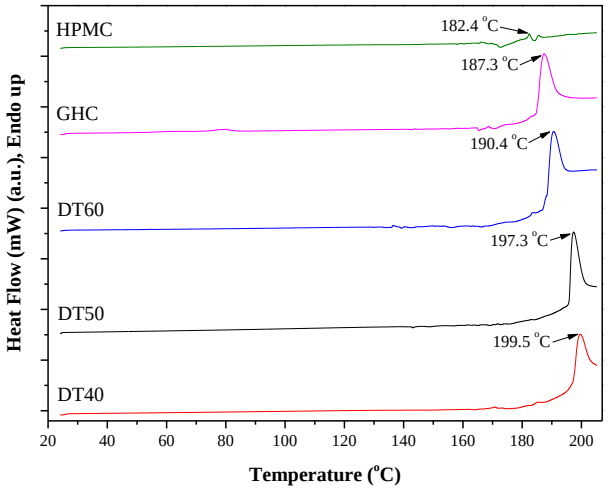

Figure 3. First heating of DSC thermogram of carrageenan based biocomposite hard capsule, gelatine and HPMC hard capsules

Additionally, an increment of drying time reduces the enthalpy of the carrageenan biocomposite hard capsule as shown in Table 3. The enthalpy of the sample reduced from 2508.58 J to 1789.28 J as the drying time increase from 40 to 60 minutes. As the drying time increased, the interaction between the carrageenan, NC, crosslinker, and toughening agent in the biocomposite through van der Walls interaction and hydrogen bonding were also decreasing [38]. Lower energy was required to break the molecular bonding in the biocomposite as the drying time increase. The enthalpy and melting point of carrageenan biocomposite was higher than the commercial gelatin and HPMC hard capsules. It proved that the developed hard capsule was more stable than the commercial hard capsules.

Table 3. Melting Temperature and enthalpy of carrageenan based biocomposite hard capsule, gelatin and HPMC hard capsules

Sample	Melting Temperature, Tm (°C)	Enthalpy, ΔH (J)
DT40	199.5	2508.58
DT50	197.3	1918.54

DT60	190.4	1789.28
GHC	187.3	1870.27
HPMC	182.4	1284.27

4. Conclusion

Drying time of biocomposite is a crucial parameter as it gives a significant effect on the shelf life and stability of the product. Drying of carrageenan based biocomposite hard capsule up to 60 minutes decreased the moisture content up to 25%. Meanwhile, the capsule loop strength and capsule hardness were also reduced as the drying time increase up to 23% and 35%, respectively. Additionally, the decrease in the moisture content and mechanical strength of hard capsule were proven by the decrease in the absorbance area of OH group in the functional group determination analysis. Reduction of the mechanical properties was also proven by the reduction of melting temperature and the enthalpy of the carrageenan based biocomposite hard capsule as the drying time increased from 40 to 60 minutes. Prolong drying time caused more water loss to the surrounding that leads to the occurrence of hydrogen bonding intermolecular interaction reduction, fragmentation of the carrageenan molecule, and case hardening phenomena in the hard capsule structure. Moreover, longer drying time led to the physicochemical and mechanical properties reduction of the carrageenan based biocomposite hard capsule. The best drying time for carrageenan biocomposite hard capsule is 40 minutes which showed the highest mechanical strength and thermal stability.

References

[1] Laha A, Sharma CS, Majumdar S (2016) Electrospun gelatin nanofibers as drug carrier: Effect of crosslinking on sustained release. Mater Today Proc 3:3484–3491 . doi: 10.1016/j.matpr.2016.10.031

[2] Abu Bakar SH, Adam F (2017) Determination of physical crosslink between carrageenan and glyoxylic acid using density functional theory calculations. Malaysian J Anal Sci 21:979–985 . doi: https://doi.org/10.17576/mjas-2017-2104-25

[3] Hamdan MA, Lakashmi SS, Mohd Amin KN, Adam F (2020) Carrageenan-based hard capsule properties at different drying time. IOP Conf Ser Mater Sci Eng 736:052005 . doi: 10.1088/1757-899X/736/5/052005

[4] Adam F, Hamdan MA, Abu Bakar SH (2020) Carrageenan; A Novel and Future Biopolymer. In: Mamun A Al, Jonathan YC (eds) Industrial Applications of Biopolymers and their Environmental Impact. Taylor & Francis, pp 224–239

[5] Arpitha GR, Yogesha B (2017) An Overview on Mechanical Property Evaluation of Natural Fiber Reinforced Polymers. Mater Today Proc 4:2755–2760 . doi: 10.1016/j.matpr.2017.02.153

[6] Mounika M, Ravindra K (2015) Characterization of Nanocomposites Reinforced with Cellulose Whiskers: A Review. Mater Today Proc 2:3610–3618 . doi: 10.1016/j.matpr.2015.07.107

[7] Ooi SY, Ahmad I, Mohd Amin MCI (2015) Effect of Cellulose Nanocrystals Content and pH on Swelling Behaviour of Gelatin Based Hydrogel. Sains Malaysiana 44:793–799

[8] Milanez DH, Amaral RM Do, Faria LIL De, Gregolin JAR (2013) Assessing nanocellulose developments using science and technology indicators. Mater Res 16:635–641 . doi: 10.1590/S1516-14392013005000033

[9] Santana PA, de Carvalho Lopes D, Neto AJS (2019) Economic analysis of low-temperature grain drying. Emirates J Food Agric 31:930–936 . doi: 10.9755/ejfa.2019.v31.i12.2040

[10] Doymaz İ, Sahin M (2016) Effect of temperature and pre-treatment on drying and rehydration characteristics of broccoli slices. J Food Meas Charact 10:364–373 . doi: 10.1007/s11694-016-9315-2

[11] Doymaz İ, Karasu S, Baslar M (2016) Effects of infrared heating on drying kinetics, antioxidant activity, phenolic content, and color of jujube fruit. J Food Meas Charact 10:283–291 . doi: 10.1007/s11694-016-9305-4
[12] Brasiello A, Adiletta G, Russo P, Crescitelli S, Albanese D, Di Matteo M (2013) Mathematical modeling of eggplant drying: Shrinkage effect. J Food Eng 114:99–105 . doi: 10.1016/j.jfoodeng.2012.07.031
[13] Nadian MH, Rafiee S, Golzarian MR (2016) Real-time monitoring of color variations of apple slices and effects of pre-treatment and drying air temperature. J Food Meas Charact 10:493–506 . doi: 10.1007/s11694-016-9328-x
[14] Cárdenas-Bailón F, Pérez-Vázquez C, Osorio-Revilla G, Gallardo-Velázquez T (2019) Shrinkage modeling, drying kinetics and quality assessment of carrot cubes dried in a two stage spouted bed drying process. Emirates J Food Agric 31:654–665 . doi: 10.9755/EJFA.2019.V31.I9.1990
[15] Amin KNM, Amiralian N, Annamalai PK, Edwards G, Chaleat C, Martin DJ, Mohd Amin KN, Amiralian N, Annamalai PK, Edwards G, Chaleat C, Martin DJ (2016) Scalable processing of thermoplastic polyurethane nanocomposites toughened with nanocellulose. Chem Eng J 302:406–416 . doi: 10.1016/j.cej.2016.05.067
[16] Hamdan MA, Adam F, Mohd Amin KN (2018) Investigation of Mixing Time on Carrageenan-Cellulose Nanocrystals (CNC) Hard Capsule for Drug Delivery Carrier
[17] Rhim JW, Wang LF (2014) Preparation and characterization of carrageenan-based nanocomposite films reinforced with clay mineral and silver nanoparticles. Appl Clay Sci 97–98:174–181 . doi: 10.1016/j.clay.2014.05.025
[18] Adam F, Hamdan MA, Hana S, Bakar A, Yusoff M, Jose R (2020) Molecular recognition of isovanillin crosslinked carrageenan biocomposite for drug delivery application for drug delivery application. Chem Eng Commun 0:1–12 . doi: 10.1080/00986445.2020.1731802
[19] Mei X, Etzler FM, Wang Z (2006) Use of texture analysis to study hydrophilic solvent effects on the mechanical properties of hard gelatin capsules. Int J Pharm 324:128–135 . doi: 10.1016/j.ijpharm.2006.06.017
[20] Hundal J, Takhar PS (2010) Experimental study on the effect of glass transition on moisture profiles and stress-crack formation during continuous and time-varying drying of maize kernels. Biosyst Eng. doi: 10.1016/j.biosystemseng.2010.03.004
[21] Ayub M, Wahab S, Durrani Y (2003) Effect of Water Activity (A w) Moisture Content and Total Microbial Count on the Overall Quality of Bread. Int J Agric Biol 8530:5–3
[22] Yeager JG, Ward RL (1981) Effects of moisture content on long-term survival and regrowth of bacteria in wastewater sludge. Appl Environ Microbiol 41:1117–1122 . doi: 10.1128/aem.41.5.1117-1122.1981
[23] Rezaei F, vander Gheynst JS (2010) Critical moisture content for microbial growth in dried food-processing residues. J Sci Food Agric 90:2000–2005 . doi: 10.1002/jsfa.4044
[24] Sederavičiūtė F, Domskienė J, Baltina I (2019) Influence of drying temperature on tensile and bursting strength of bacterial cellulose biofilm. Mater Sci 25:316–321 . doi: 10.5755/j01.ms.25.3.20764
[25] Bal LM, Yogranjan, Naik SN, Satya S, Kar A (2017) Changes in tissue structure and physico-chemical quality characteristics of bamboo shoot slices during microwave drying process. J Food Meas Charact 11:1203–1209 . doi: 10.1007/s11694-017-9497-2
[26] Bakhara CK, Pal US, Bal LM (2018) Drying characteristic and physico-chemical evaluation of tender jackfruit slices during osmo-convective drying. J Food Meas Charact 12:564–572 . doi: 10.1007/s11694-017-9668-1
[27] Lilia ML, Juan RR, David RV, Anabel LO (2017) Changes in physical properties and relations with allicin degradation during convective drying of garlic. J Food Meas Charact 11:1227–1232 . doi: 10.1007/s11694-017-9499-0
[28] Maskan M (2000) Microwave/air and microwave finish drying of banana. J Food Eng 44:71–

78
[29] Bodirlau R, Teaca CA, Spiridon I (2013) Influence of natural fillers on the properties of starch-based biocomposite films. Compos Part B Eng 44:575–583 . doi: 10.1016/j.compositesb.2012.02.039
[30] Dewi EN, Darmanto Y, Ambariyanto (2012) Characterization and quality of semi refined carrageenan (SCR) products from different coastal waters based on fourier transform infrared technique. J Coast Dev 16:25–31 . doi: 10.4236/jbnb.2011.225070
[31] Hosseinzadeh H (2009) Potassium persulfate induced grafting of polyacrylamide onto kappa-carrageenan. J Appl Chem 4:9–20
[32] Uddin M, Mamun A, Rashid M, Asaduzzaman M (2016) In-process and Finished Products Quality Control Tests for Pharmaceutical Capsules According to Pharmacopoeias. Br J Pharm Res 9:1–9 . doi: 10.9734/BJPR/2016/22044
[33] Paglione I dos S, Galindo MV, de Souza KC, Yamashita F, Grosso CRF, Sakanaka LS, Shirai MA (2019) Optimization of the conditions for producing soy protein isolate films. Emirates J Food Agric 31:297–303 . doi: 10.9755/ejfa.2019.v31.i4.1933
[34] Thommes M, Blaschek W, Kleinebudde P (2007) Effect of drying on extruded pellets based on κ-carrageenan. Eur J Pharm Sci 31:112–118 . doi: 10.1016/j.ejps.2007.02.010
[35] Satish G, Prasad VVS, Ramji K (2018) Effect on Mechanical Properties of Carbon Nanotube Based Composite. Mater Today Proc 5:7725–7734 . doi: 10.1016/j.matpr.2017.11.449
[36] López-Ortiz A, Rodríguez-Ramírez J, Méndez-Lagunas LL (2013) Effects of drying air temperature on the structural properties of garlic evaluated during drying. Int J Food Prop 16:1516–1529 . doi: 10.1080/10942912.2011.599090
[37] Rochas C, Rinaudo M (1980) Activity Coefficients of Counterions and Conformation in Kappa-Carrageenan Systems. Biopolymers 19:1675–1687
[38] Mohd Amin KN (2016) Cellulose nanocrystals reinforced thermoplastic polyurethane nanocomposites. The University of Queensland

Acknowledgement
We acknowledge financial support from UMP Postgraduate Research Grants Scheme (PGRS1903130) for project funding.