Utilization of swimming crab by-product as a seafood flavor microcapsules obtained by spray drying

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Abstract. Utilization of swimming crab by-products is not optimal, so it is proposed to produce a powdered flavor enhancer considering that the by-product of crab is rich in umami compounds. Micro-encapsulation technology can be used to achieve this purpose, thus its application in the food sector becomes broader. In this study, seafood flavor enhancer products were produced using a mixture of maltodextrin (MD) and arabic gum (AG) as coating materials in a ratio of 2:1, 1:1 and 1:2 under spray drying. The results showed that the seafood flavor microcapsules (SFM) obtained had a low water content and water activity values, the particles had a bright color and were classified as hygroscopic, with a high solubility, a fast curing time and the particles had a good flowability (especially MD:AG ratio 1:2). The results of the transmission electron microscopy showed that increasing the AG ratio resulted in particles with a smooth, spherical shape without holes or pores. SFM in the 1:2 MD:AG ratiogroup had the lowest particle size, with the highest L-glutamic acid content. It was concluded that an MD:AG ratio of 1:2 produced the SFM with the best physical and chemical characteristics.
Key Words: seafood flavor microcapsules, spray dryer, maltodextrin, arabic gum.

Introduction. Swimming crab is one of the leading fishery products in Indonesia. Until the end of 2020, it was reported that the export volume of the Indonesian swimming crab-crab group reached 30.8 thousand tons (MMAF 2021). The crab industry certainly produces waste, both in solid and liquid form. Crab solid waste consists of 55% shell and 5% body reject. Meanwhile, liquid waste reaches 25% (Sasongko et al 2018). Crab shells are reported to have a glutamic acid content of 1,150 mg 100 g^{-1} (Yonata et al 2021), as well as lemi, which is a crab solid waste containing 15.65% protein as a source of glutamic amino acid. Compounds L-glutamic acid and its salts, as well as monosodium glutamate (MSG), are the main compounds forming the umami taste in food. Discovered by Ikeda in 1908, umami taste is described as a delicious or savory taste which is the fifth basic taste (Yamaguchi & Ninomiya 2000).

Umami compounds from crab by-products can be obtained through the heating process. The protein hydrolysis process with hot water will produce free amino acids, including the compound L-glutamic acid (Harada-Padermo et al 2020). The protein hydrolyzate, similarly to the protein peptides, has attracted the interest of many researchers, as a source of umami compounds. Umami has a very fast release during a high-temperature process, so microencapsulation technology can be used to control the release of umami compounds. In addition, microcapsules are easier to apply in the food sector. More specifically, spray drying is a processing method that has been widely recommended for producing umami flavor enhancer powders (Kanpairo et al 2012; Cho et al 2015; Mayasari et al 2020; Harada-Padermo et al 2020). There are many factors that influence the success of the microencapsulation of umami compounds using the spray dryer method. Wang & Selomulya (2020) have summarized that the coating material is one of the most important factors that will affect the characteristics of the resulting microcapsules. Recent studies have reported that maltodextrin (MD), arabic gum (AG) and chitosan, as coating materials for umami compounds, can control the release time during a high-temperature process (Bu et al 2021). Mixed of MD-AG coating materials resulted in MSG microcapsules with excellent thermal stability. MSG microcapsules coated with MD-AG had a controlled release time of up to 60 minutes during boiling in hot water. In addition, the FTIR results also demonstrated the successful encapsulation of MSG by MD-AG during spray drying (Wu et al 2019). Based on the information, the mixture of MD-AG has a great potential to be used as a coating material in producing seafood flavor microcapsules (SFM) from swimming crab by-products, using the spray drying method. The right MD-AG ratio is expected to produce SFM with characteristics that are preferred by consumers. This study aims to determine the best MD:AG ratio in producing SFM from swimming crab by-product using the spray drying method. Parameters analyzed include L-glutamic acid content, moisture content, water activity, hygroscopicity, solubility, wettability, bulk and tapped density, compressibility index, Hausner ratio, flowability, color characteristic, morphology and particle size distribution.

Material and Method. Swimming crab by-products are obtained from the crab processing industry (CV Minajaya) located in Semarang, Indonesia. The coating materials used were MD (DE 9-13) and AG obtained from distributors in Semarang. The by-products of crab shells and lemi were initially cleaned of dirt, then dried using oven drying (Agroindo OVG-12) at 60°C for 6 hours. Then, they were crushed using a disc mill (Agroindo AGC-15) to obtain crab by-product flour (FSCB) measuring 100 mesh. The extraction process of umami compounds from swimming crab by-products refers to the research of Poojary et al (2017): as much as 20 g of FSCB was immersed in 1,000 mL of distilled water in a screw-top flask and stirred in a shaken water bath at a controlled temperature of 70°C for 30 minutes. After the extraction was completed, the sample was filtered through a Whatman filter paper of size 41, under vacuum, in order to obtain a seafood flavor enhancer extract (SFEE).

The microencapsulation process uses the spray drying method. 500 mL of SFEE was added as a coating material in the form of a mixture of 30% (w/v) MD-AG (2:1, 1:1 and 1:2). It was then mixed for 15 minutes using a homogenizer (Daihan HG-15D) at high speed (3000 rpm) and dried by spray drying (Buchi, B-190) at an inlet temperature of 120±2°C, an outlet temperature of 80±2°C and the rate of feed flow of 6.0 mL min^{-1}, at 1.5 bar pressure (Nurhidajah et al 2022). The SFM obtained was then analyzed for quality characteristics in the form of moisture content (Shimadzu, MOC63u), water activity (Rotronic, Hygropalm-HP23-Aw-A), hygroscopicity (Caparino et al 2012), solubility (Vidović et al 2014), wettability (Gong et al 2008), bulk and tapped density (Wang et al 2019), compressibility index and Hausner ratio (Lebrun et al 2012), flowability, color characteristic (Minolta CR-310 Chromameter) (Caparino et al 2012), morphology (SEM, JSM-6510 LA), particle size distribution (LPSA, LLPA-C10), and L-glutamic acid (L-glutamic acid assay kit, Megazyme).

***Statistical analysis**.* All data obtained were analyzed using a one-way ANOVA test and followed by a post hoc LSD test to determine significant differences between the mean variables for the selected parameters. The significance level of the differences was defined at $p<0.05$. Statistical analyzes were performed using SPSS 22.0 software.

Results and Discussion

Microencapsulation yield and L-glutamic acid content. SFM spray drying was performed using a mixture of MD:AG, as a coating material, yielding about 39.02 to 47.38 g 100 g^{-1} SFM, a result which is considered quite good for a laboratory scale. However, this result is still lower than în the study of Harada-Padermo et al (2020) for spray drying of mushroom extracts using MD coating material. Yield is strongly influenced by the amount of material deposited on the walls of the equipment, aș droplets and powder, that can stick to the walls and cyclones, reducing the number of particles that can be collected at the end of the process (Wang & Langrish 2009). It can be seen that an increase in the AG ratio has an impact, causing a significant decrease in the SFM yield; this result is in line with the research of Cid-Ortega & Guerrero-Beltrán (2020).

This is due to the short branched structure of AG and its high hydrophilic nature, which allows many particles to adhere to the walls of the spray dryer (Tonon et al 2009). However, the levels of L-glutamic acid SFM increased significantly (885.03 to 911.01 mg 100 g^{-1}) as the AG ratio increased. The ability of AG to form films is very high, so the core material will be trapped more (Mahdi et al 2020). The MD:AG ratio of 1:1 showed competitive results regarding the yield and levels of L-glutamic acid SFM, compared to other treatments.

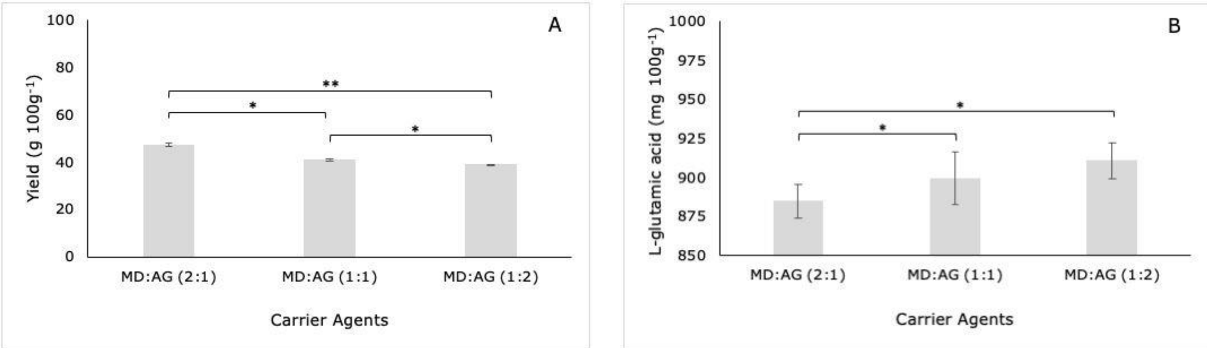


Figure 1. Yield (A), and L-glutamic content of seafood flavor microcapsules.

Moisture content and water activity. The moisture content and water activity of SFM can be seen in Table 1. In general, the water content of SFM ranges from 2.24 to 3.28%. Moisture content below 5% is categorized as good enough so that it is safe to store (Manickavasagan et al 2015). Regarding the effect of the MD:AG ratio, there is a tendency to increase the water content of SFM when the AG ratio is added more to the coating mixture. However, a significant increase in water content was only seen when the MD:AG ratio was 1:2. This result is in line with the research of Lourenço et al (2020), stating that AG significantly produces particles with a higher moisture content than the MD. AG has complex hetero-polysaccharide properties and a branched structure, containing shorter chains and more hydrophilic groups (Bazaria & Kumar 2017). Thus, AG has the ability to absorb water from the surrounding environment, more than MD, as has been reported by Tran & Nguyen (2018). SFM has a low water activity and there is no significant difference between treatments. As depicted in Table 1, the water activity of the SFM ranged from 0.23 to 0.25. This value indicates that SFM remains stable under the microbial contamination action, with a water activity lower than 0.6. In addition to inhibiting microbial growth, the low water activity will overcome the coagulation problems and will increase the physicochemical stability and total acceptability (Chew et al 2018).

Hygroscopicity. Hygroscopicity is the ability of a material to absorb moisture from the environment. High hygroscopicity will cause aggregation of spray dryer powder, so it will affect the nutrition and flow properties of the powder. The hygroscopicity of the SFM produced with a 1:2 ratio MD:AG (18.57%) was significantly higher than for the SFM produced with a 1:1 ratio MD:AG (15.02%) and a 2:1 ratio MD:AG (14.41%). Similar results were also reported by Manickavasagan et al (2015): AG, as a coating material, will produce a more hygroscopic powder than MD. This is because MD has anti-hygroscopic properties which can change the balance of hydrophilic/hydrophobic particles, which then reduces the rate of water absorption from the environment (Wang et al 2011). The phenomenon of increasing hygroscopicity of SFM along with the increasing presence of AG may be due to the higher moisture adsorption of AG, which is then associated with the relationship between the hydrogen present in the water molecule and the available hydroxyl groups In the amorphous region of the substrate, as well as in the surface of the crystalline region (Tonon et al 2009). The highly branched AG structure and size small particles will produce a hygroscopic powder (Du et al 2014).

Table 1
Moisture content, water activity, and physical properties of seafood flavor microcapsules

Analysis	*Carrier agents (MD:AG)*		
	2:1	*1:1*	*1:2*
Moisture content (%)	2.24±0.11[a]	2.37±0.08[a]	3.28±0.09[b]
Water activity	0.25±0.01[a]	0.23±0.02[a]	0.24±0.02[a]
Hygroscopicity (%)	14.41±0.38[a]	15.02±0.19[a]	18.57±0.41[b]
Solubility (%)	99.86±0.88[a]	98.53±0.43[a]	97.84±0.94[a]
Wettability (min)	5.53±0.79[a]	6.45±0.97[b]	9.23±0.96[c]
Bulk density (g cm^{-3})	0.410±0.14[a]	0.473±0.28[b]	0.494±0.11[c]
Tapped density (g cm^{-3})	0.515±0.71[a]	0.569±0.36[b]	0.580±0.29[c]
Compressibility index (%)	25.55±0.74[a]	18.25±1.85[b]	14.03±1.48[c]
Hausner ratio	1.26±0.01[a]	1.20±0.02[b]	1.17±0.02[c]
Flowability	Passable	Fair	Good
L*	95.68±0.79[c]	93.49±0.52[b]	92.11±0.60[a]
a*	0.31±0.01[a]	0.29±0.02[a]	0.30±0.01[a]
b*	4.43±0.14[a]	4.28±0.16[a]	4.29±0.09[a]

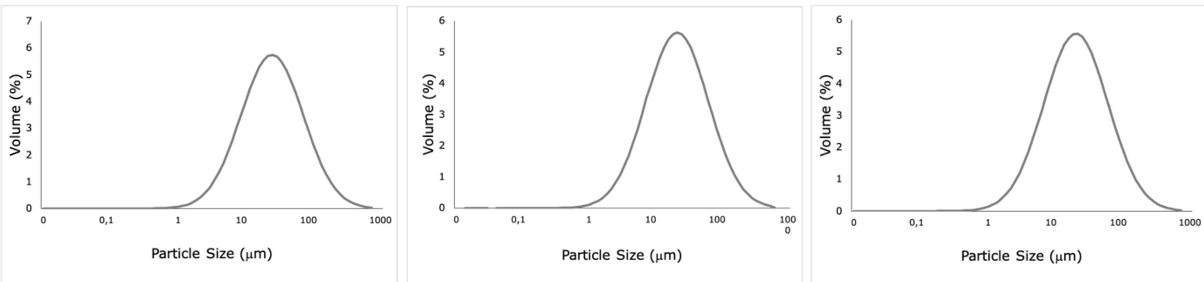
Values are the mean ± standard deviation of quintuplicate treatments. Different superscripts in the same row showed statistically significant differences ($p<0.05$) as determined by LSD.

Solubility and wettability. Solubility determines the particle dissolution capability and is a decisive factor for the quality of the powders used as ingredients in the food industry (Mahdi et al 2019). Powders with a low solubility will affect the level of consumer acceptance. The average solubility value of SFM, as listed in Table 1, ranged from 97.84 to 98.86%, slightly lower than the solubility of the flavor enhancer from the shiitake by-products, 99.03%, asreported by Harada-Padermo et al (2020). There was no significant difference between the ratio of the MD:AG used to the resulting SFM dissolution, but the SFM solubility tended to be higher when the MD concentration was increased. Du et al (2014) previously reported that spray-dried powders produced with MD had better solubility, compared to those produced with AG. This is because MD has a hydroxyl group (OH) in its molecule (Avila et al 2015). Solubility is also associated with the powder particle structure: the MD generally produces particles with a more amorphous surface and larger cavity size, thus powders will be more soluble in water (Mayasari et al 2020). In addition to a high solubility, the powder is also expected to have a fast wettability time. Wettability itself is the ability of powder to absorb water associated with powder reconstitution. The shorter the dissolving time of the powder into water, the better its physical properties in food processing (Chew et al 2018). As observed in Table 1, the SFM produced with 1:1 MD:AG showed the lowest wettability (5.53 min), significantly different from the other treatments. Ferrari et al (2012) also previously reported a shorter wettability time for the microcapsules than for the AG. Wettability and solubility are highly dependent on the surface area and particle size. Powders with a larger particle size have a higher void volume, tending to be more permeable to water. On the other hand, the smaller particles are less porous, so it is more difficult for the liquid to penetrate into the particle matrix (Lourenço et al 2020).

Bulk density, tapped density, compressibility index, Hausner ratio and flowability. Bulk and tapped densities are parameters used to determine the weight and amount of material to be accommodated in a container: a dry product will be denser and the storage process will be more efficient (Fernandes et al 2014). The value of SFM bulk density ranged from 0.410 to 0.494 g cm^{-3}, while the value of tapped density SFM was 0.515 to 0.580 g cm^{-3}. Increasing the MD ratio results in lower bulk and tapped density SFM values. Lower bulk and tapped density values tend to contain more air, so the possibility of product oxidation is higher (Lourenco et al 2020). Some literature also relates that a low water content will produce powders with low bulk and tapped density values (Nadeem et al 2011).

Particle size distribution is a factor that can affect the bulk and tapped density. It is expected that powders with lower particle sizes will produce higher bulk and tapped densities. In this study, the particle size tends to be smaller when produced with the predominant AG coating material (Figure 2C and Table 2). In addition, the SFM formed had a completely spherical structure without any hollows (Figure 3C), while the SFM produced with an MD:AG ratio of 2:1 and a ratio of 1:1 had a more hollow structure, there were hollows and imperfect circles (Figures 3A and 3B). As the particle structure becomes hollower, the particle volume increases, which affects its cohesiveness. Good cohesiveness is indicated by low Hausner ratio (HR) and compressibility index (CI) values, and correlates with the flowability of a particle. It is expected that the SFM produced has a value of HR<1.34 and CI<25%, thus the particle flowability will be classified as good (Lebrun et al 2012). Based on the data from Table 1, the SFM produced with various ratios of MD:AG had quite good HR (1.17 to 1.26) and CI (14.03 to 25.55%) values. The MD:AG ratio of 1:2 is recommended because it produces SFM with the best flowability ("good" category).

Color parameters. The color attribute is one of the sensory attractions that need attention. Ideally, umami flavor enhancers are expected to have bright color characteristics so that they do not affect the final color of the product when added. The FSCB used has a fairly bright color characteristic (L* 77.02) slightly yellowish (a* 0.49 and b* 11.73, as defined in the CIELAB system, where L* stands for the perceptual lightness, and a* and b* are parameters quantifying the four unique colors of the human vision: red, green, blue and yellow). MD:AG resulted in SFM with a very bright color where the L* value ranged from 92.11 to 95.68, this is better than the flavor enhancer from shiitake byproducts, reported by Harada-Padermo et al (2020). The brightness of SFM decreased with increasing the AG ratio, while the values of a* (0.29 to 0.31) and b* (4.28 to 4.23) of the SFM tended to be the same, no significant difference was seen (Table 1). Indirectly, the difference in SFM brightness is caused by the L* value of each coating material. MD in this study has a higher brightness (L*=98.39) than AG (L*=91.95), so a higher MD ratio will result in a brighter SFM. The product under study showed positive results, as dark colored ingredients have limited application in food (Toledo et al 2019).



(A) (B) (C)

Figure 2. Particle size distribution images of seafood flavor microcapsules produced at various ratios MD:AG, (A) 2:1; (B) 1:1, and (C) 1:2.

Particle size distributions. Particle size distribution is an important parameter of the quality of a particulate system because it affects transportation, storage, as well as physical and chemical properties, changing its performance (Tontul & Topuz 2017). D10, D50, and D90, representing 10, 50, and 90% of the volumetric diameter of the accumulated particles, respectively, presented a unimodal distribution (Figures 2A, 2B, and 2C). D[4,3] is the value where the average diameter of particles that have the same mass to particle volume ratio, or is commonly used to indicate the average diameter of particle size. SFM produced with an MD:AG ratio of 2:1 has particles with the largest average diameter, which is 44.371 μm, followed by an MG:AG ratio of 1:1, with 39.826 μm, the smallest particles being produced with an MD:AG ratio of 1:2, with a diameter of

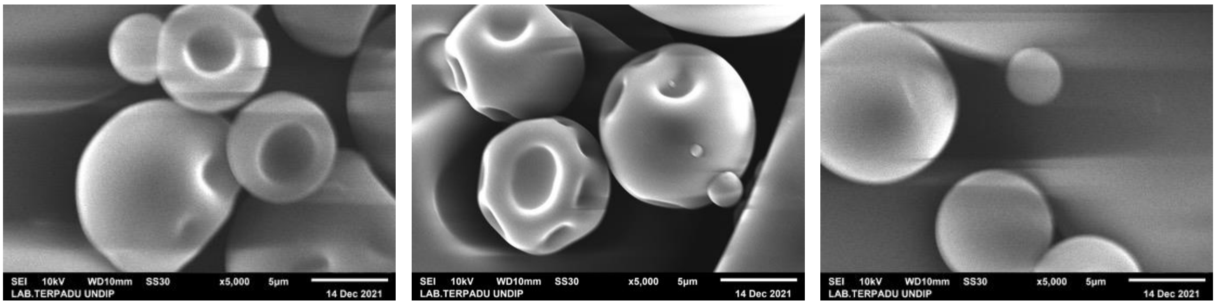
37.726 μm. Based on these data, it was concluded that an increasing MD ratio in the coating material would result in SFM with larger particles. Our results are similar to those reported by Du et al (2014). The particle size has a negative correlation with the bulk density of the powder. Particles with smaller sizes generally have a larger bulk density. This is because the smaller the average diameter of the particles, the lower the interstitial air content between the particles, so that there is less free space left because the particles are already occupied (Goula & Adamopoulos 2010). However, particles with a small diameter tend to require a longer wetting time because of the lower particle porosity.

Table 2

Particle size distribution parameters of seafood flavor microcapsules

Carrier agents (MD:AG)	*D [4,3] (μm)*	*D [3,2] (μm)*	*D 10 (μm)*	*D 50 (μm)*	*D 90 (μm)*
2:1	44.371	14.428	6.152	25.320	98.394
1:1	39.826	12.372	5.559	22.213	88.740
1:2	37.726	11.402	5.110	20.754	84.256

Morphology and particle size distributions. Morphological images of SFM are shown in Figure 3 (3A, 3B and 3C). SFM produced with MD:AG ratios of 2:1 and 1:1 are relatively similar: the resulting particles have a smooth surface, but are not perfectly round, many surfaces looking dented. However, SFM produced with an MD:AG ratio of 1:2 produces particles that are smooth and perfectly round, with no visible agglomeration or surface dents. This result has previously been reported by Cano-Chauca et al (2005): spray dried mango powder produced with MD has a more amorphous structure, aggregated, with non-uniform size. However, powders produced with AG will result in a more uniform structure form, with a very smooth and intact surface.



(A) (B) (C)

Figure 3. Morphology images of seafood flavor microcapsules produced at various ratios MD:AG, (A) 2:1; (B) 1:1, and (C) 1:2.

According to Saénz et al (2009), surface dents are formed due to a shrinkage of the particles during drying and cooling, and the presence of these dents hurts the flow properties of powder particles. It can be seen that the HR and CI values of SFM will be better when the surface of the spherical particles is intact (Table 1). Microcapsules with a a smooth surface indicate that the temperature used for drying is very precise (Li et al 2011). It has also been reported that the success of the microencapsulation of umami compounds by spray drying is characterized by the smooth surface of the microcapsules and an intact spherical shape (Wu et al 2019). AG is known to have a very high film-forming ability (Mahdi et al 2019), this indicates that the higher the concentration of AG, the more umami source compounds are successfully encapsulated.

Conclusions. Based on the analysis of physical and chemical properties, the best SFM resulted from the swimming crab by-product produced with an MD:AG ratio of 1:2. The production of SFM with an MD:AG ratio of 1:2, under the spray drying, resulted in a yield of 39.02 g 100 g^{-1}, with L-glutamic acid content reaching 911.01 mg 100 g^{-1}. SFM is known to have a low water content, of 3.28%, a water activity of 0.24 and hygroscopicity of 18.57%, a high solubility, of 97.84%, and a fairly good wettability time, of 9.23 min. SFM has a very bright color characteristics (L* 92.11; a* 0.30; b* 4.29), a good flowability and particle cohesiveness, as indicated by their bulk and tapped densities (of 0.494 g cm^{-3} and 0.580 g cm^{-3}, respectively), and by their CI and HR values (of 14.03 and 1.17, respectively). SFM has an average particle size of 37.726 μm. The results of the transmission electron microscopy showed that the SFM had a smooth particle surface and a spherical shape, without holes or pores.

Conflict of interest. The authors declare no conflict of interest.

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Received: 04 January 2022. Accepted: 15 March 2022. Published online: 28 March 2022.
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How to cite this article:
Yusuf M., Yonata D., Pranata B., Nurhidajah, 2022 Utilization of swimming crab by-product as a seafood flavor microcapsules obtained by spray drying. AACL Bioflux 15(2):716-724.