

Turnitin-Microencapsulation of seafood flavor enhancers from Indonesian brown seaweed with maltodextrin, Arabic gum, and β -cyclodextrin

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Microencapsulation of seafood flavor enhancers from Indonesian brown seaweed with maltodextrin, Arabic gum, and β -cyclodextrin

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ABSTRACT

This study aimed to improve the physical and chemical properties of seafood flavour enhancer microcapsules from *Sargassum aquifolium* extract using a combination of maltodextrin (MDE), Arabic gum (AG) and β -cyclodextrin (β CD) as coating materials. Therefore, this study used MDE-AG, MDE-AG- β CD and MDE- β CD as coating materials for the water extract of *Sargassum aquifolium* as a source of flavour using the spray drying method. The results showed that there was a significant increase in the physical and chemical properties of the microcapsules. MDE-AG- β CD coating material produced the highest product yield (42.18%), and 2-glutamic acid content was 0.55 g/100g. Besides that, it also improves the water content (2.61%), water activity (0.40), and hygroscopicity (8.24%) of the microcapsules. Although the solubility (95.54%) and colour attribute (darker) of microcapsules decreased, the microcapsules obtained had a perfectly spherical particle shape with a smooth surface and a smaller particle size (2.304 μ m). In conclusion, MDE-AG- β CD coating material is highly recommended for seafood flavour enhancer microcapsules.

INTRODUCTION

Brown seaweed has long been used in the food industry, especially as a source of alginate. *Sargassum* sp from Indonesia is a species of brown seaweed with very abundant availability (Puspita *et al.*, 2020). *Sargassum* sp contains a high protein component, around 6.21-8.54% (Dewinta *et al.*, 2020). These proteins contain various amino acids, such as glutamic acid and aspartic acid, with high concentrations (Pratiwi *et al.*, 2021). In their free form, glutamic acid and aspartic acid produce a distinctive aroma and have been developed as umami flavour enhancers (Mouritsen *et al.*, 2012).

The umami flavour of brown seaweed has aroused the interest of researchers in recent years (Viera *et al.*, 2018; Mouritsen *et al.*, 2019; Milinovic *et al.*, 2020). Recently, brown seaweed *Sargassum aquifolium* from Indonesia was reported to have high levels of glutamic acid and aspartic acid, so it has the potential to be developed as a source of umami (Pratiwi *et al.*, 2021). Umami compounds generally have low thermal stability and quickly degrade during drying and processing (Zhou *et al.*, 2017). Spray

drying ² has been proven to increase the thermal stability of umami compounds. Adding a coating agent in the spray drying process significantly protects the umami compounds during ¹ drying (Bu *et al.*, 2021).

Coating material is one of the determining factors for the success of microencapsulation process of umami compounds (Wang and Selomulya, 2020). Maltodextrin (MDE) has been used in microencapsulation process of free amino acids from brown seaweed (Nurhidajah *et al.*, 2021), as well as Arabic gum (AG) in the microencapsulation process of umami compounds from hydrolyzed rebon shrimp protein (Suparmi *et al.*, 2021). Other researchers used a combination of MDE and AG to microencapsulate umami compounds from free amino acid extracts of crab waste (Yusuf *et al.*, 2021). However, MDE and AG is considered not optimal, the efficiency of microencapsulation and yield of product still need to be improved. In addition, microcapsules obtained tend to be hygroscopic, and the umami component in microcapsules is still easily degraded during processing (Larasati *et al.*, 2019).

Recent studies have revealed that the addition of β -cyclodextrin (β CD) in coating material formula can significantly restrain the degradation rate of flavour and aroma-producing compounds during processing and storage (Pellicer *et al.*, 2019). β CD is a cyclic oligosaccharide that has excellent thermal stability. The structure of β CD is unique because it allows the formation of inclusion complexes that trap all or part of the guest molecules in their hydrophobic cavities, mainly through hydrogen bonds (Reineccius *et al.*, 2002; Charoenlap *et al.*, 2004). It is just that the solubility of β CD in water is relatively low compared to other linear dextrans, so the ratio of adding β CD needs to be evaluated (Szente and Szejtli, 2004; Krishnaswamy *et al.*, 2012).

This research aimed to improve the physical and chemical properties of umami flavour-enhancing microcapsules from free amino acid extract *Sargassum aquifolium* using MDE, AG and β CD coating materials by spray drying method, which is considered the most feasible, economical and applicable method. For this reason, the obtained microcapsules were characterized in terms of yield, moisture content, water activity, hygroscopicity, solubility, colour properties, morphological characteristics and particle size, also L-glutamic acid content as a source of umami taste.

MATERIALS AND METHODS

² Brown seaweed species *Sargassum aquifolium* ² collected from the coast of Garut, West Java, Indonesia. Coating materials include MDE with Dextrose Equivalent (DE) 9-13 (Neo-Maldex), AG (Ingredient), and β CD (Sigma-Aldrich). Distilled water, ¹ HCl (Sigma-Aldrich) and L-glutamic acid (Megazyme). The brown seaweed was first cleaned of impurities ¹ then dried using a drying oven (Agrowindo OVG-12) at 60 °C for 6 hours, and ground using a disc mill (Agrowindo AGC-15) to obtain seaweed powder. Extracting umami compounds was started by preparing 50 g of *Sargassum aquifolium* powder, then extracting with 1000 mL of distilled water in a beaker glass. Extraction temperature was

kept constant at 70 °C using a magnetic hot plate stirrer (Ika Magnetic Stirrer C-MAG HS7). Stirring speed is 180 rpm for 30 minutes (Nurhidajah *et al.*, 2021). The extract obtained was then filtered (Whatman No. 41), and the filtrate was frozen until used.

Microencapsulation of umami flavour compounds from *Sargassum aquifolium* extract refers to previous studies (Nurhidajah *et al.*, 2021; Yusuf *et al.*, 2021) with slight modifications. *Sargassum aquifolium* extract was prepared, then MDE-AG (9:1), MDE-AG- β CD (9:0.5:0.5) and MDE- β CD (9:1) added 30% (w/v). Then it was homogenized separately using a homogenizer (Daihan HG-15D) for 15 minutes at 3000 rpm and then dried with a BUCHI Spray Dryer B-190 at an inlet air temperature of 120 \pm 5 °C, an outlet temperature of 80 \pm 5 °C, a feed flow rate of 6.0 mL/minute, and a pressure of 1.5 bar. The obtained microcapsules were then packed in a vacuum sealer and stored until analyzed.

The analysis carried out included yield, L-glutamic acid content using a kit from Megazyme (Nurhidajah *et al.*, 2021); water content using a moisture analyzer (Shimadzu MOC63u); water activity (Rotronic Hygropalm-HP23-Aw-A); hygroscopicity (Yusuf *et al.*, 2021); solubility (Vidovic *et al.*, 2014); colour characteristics using the Minolta CR-310 Chromameter (Caparino *et al.*, 2012); particle morphology using the Scanning Electron Microscopy method with the JSM-6510LA (Jeol Ltd); and particle size analysis using the LLPA-C10 (Labron Equipment Ltd).

RESULTS AND DISCUSSION

1. Product yield

Product yield is presented in Table (1). Coating material type significantly affects the yield of microcapsules (40.92 to 46.21%); the highest product yield is produced from the MDE- β CD coating material. Adding β CD in coating material formula appears to positively affect product yield, whereas the presence of AG gives the opposite result. The same results have also been reported by previous researchers (Yusuf *et al.*, 2021). This is because AG has a short chain branch structure which is hydrophilic so that it sticks more to the wall of the spray dryer (Tonon *et al.*, 2010). The increase in product yield with the addition of β CD in coating material formula was also observed by Laokuldilok *et al.* (2016). β CD has a high molecular weight, and its addition to the coating material formula causes an increase in glass transition temperature of the particles (Truong *et al.*, 2005). Under these conditions, rate of physicochemical changes in the product, such as stickiness, collapse, and clumping to agglomeration will decrease so that more coating material is available to form microcapsules, and the product yield increases (Quispe-Condori *et al.*, 2011).

2. L-Glutamic acid content

Microcapsules obtained had relatively high levels of L-glutamic acid, ranging from 0.47 to 0.51 g/100g (**Table 1**). It can be seen that the combination of MDE, AG and β CD as coating material significantly produced the highest levels of L-glutamic acid compared to MDE-AG and MDE- β CD. MDE has a chemical structure suitable for the entrapment of umami compound **1** as a core material (**Mayasari *et al.*, 2020**). AG can form excellent films; thus, more core material will be trapped (**Mahdi *et al.*, 2020**). β CD, as a coating material, will form a hydrophobic hollow structure; through its hydrogen bonds, it will trap the core compound molecularly through the inclusion complex. Furthermore, β CD with its excellent thermal stability will protect the core compound from external disturbances during the drying process (**Deng *et al.*, 2022**).

Table (1): Physicochemical characteristic of microcapsules based on various types of coating materials

Analysis	Type of coating material		
	MDE-AG	MDE-AG- β CD	MDE- β CD
Yield (%)	40.92 \pm 0.89 ^a	42.18 \pm 0.73 ^b	46.21 \pm 0.56 ^c
L-glutamic acid (g/100 g)	0.47 \pm 0.02 ^a	0.55 \pm 0.02 ^c	0.51 \pm 0.01 ^b
Moisture content (%)	2.46 \pm 0.18 ^a	2.61 \pm 0.82 ^c	2.54 \pm 0.24 ^b
Water activity	0.43 \pm 0.01 ^b	0.40 \pm 0.01 ^a	0.41 \pm 0.01 ^a
Hygroscopicity (%)	10.33 \pm 0.11 ^c	8.24 \pm 0.82 ^a	9.38 \pm 0.34 ^b
1 Solubility (%)	97.40 \pm 0.62 ^c	95.54 \pm 0.39 ^a	94.08 \pm 0.21 ^b

Values are the average \pm standard deviation of 5 replicates. Different superscripts in the same line show statistically significant differences ($p < 0.05$), as determined by the LSD test.

3. **1** Moisture Content and Water Activity

Moisture content and water activity of the microcapsule can be seen in **Table (1)**. It was concluded that the type of coating material had a significant effect on Moisture content and water activity of the product. Moisture content of the microcapsules ranged from 2.46 to 2.61%. The presence of β CD in the coating material formulation showed a significant increase in water content. The same result was also reported by **Pasrija *et al.* (2015)**, where there was an increase in the water content of green tea extract microcapsules with the addition of β CD. This is because the β CD molecules form hydrophobic cavities to trap the core material so that **6** some of the water molecules are difficult to evaporate during the drying process (**Zhu *et al.*, 2019**). According to **Manickavasagan *et al.* (2015)**, products with a moisture content below 5.0% meet commercial powder products' requirements because they tend to be safe for storage. Other researchers suggested that the moisture content of spray-dried powder is expected to be in the range of 4-5% for storage stability (**Ghandi *et al.*, 2012**). Microcapsules with too low a moisture content tend to have a high water activity. As seen in **Table (1)**, the water activity of microcapsules tends to increase (0.40-0.43) when the product's water content is lower. However, the value of the water activity of the microcapsules in this

study was still quite good. The threshold for the activity value of dry powder product water is 0.6. This is related to the problem of coagulation, physicochemical stability, and total acceptability of the product to be produced. Besides that, microbial contamination problems may also occur when the water activity value of the product is too high (Chew *et al.*, 2018).

4. Hygroscopicity and Solubility

Microcapsules had a hygroscopicity value of 8.24⁴ to 10.33, with a solubility of 94.08 to 97.40% (Table 1). Microcapsules hygroscopicity in this study was lower than in previous studies using MDE as a coating material (Nurhidajah *et al.*, 2021). Chew *et al.* (2018) also observed a decrease in the hygroscopicity of kenaf seed oil microcapsules after being coated with AG and β CD. The decrease in microcapsule hygroscopicity is associated with the limited presence of hydrogen and hydroxyl groups in AG and β CD molecules (Fernandes *et al.*, 2014). Unlike hygroscopicity, solubility of microcapsules was seen to decrease significantly with the addition of AG and β CD. In contrast, the presence of MDE will increase product solubility. This is because the MDE structure has a very high hydroxyl group (Avila *et al.*, 2015). In addition, solubility of β CD in water at room temperature is relatively low. β CD structure is hydrophobic and the hydrogen bonds between the hydroxyl groups will cause the structure to become rigid (Szente and Szejtli, 2004; Krishnaswamy *et al.*, 2012).

5. Color Properties

Colour attribute is one of the crucial attributes in food products because it is related to the sensory appeal. Colour coordinates of microcapsules can be seen in Table (2). Microcapsules have a bright colour where the L (lightness) value ranges from 68.45-70.86. The brightness of microcapsules is strongly influenced by the L value of the coating material. BCD, MDE, and AG have L values of 99.28, 98.39 and 91.95 (data not shown). The hue and chroma values represent the colour angle and colour intensity. The hue value of the resulting microcapsules ranges from 87.26 – 87.79°, so the microcapsules tend to form a yellow colour (Hutching, 1999). The colour intensity of the microcapsules is relatively low (8.81-9.83); thus, the colour of the microcapsules produced is faded yellow. Compared to previous studies, the microcapsules in this study tend to have darker colour characteristics (Nurhidajah *et al.*, 2021). However, it appears that the combination of various coatings used did not affect the colour attributes of the microcapsules.

Table (2): Color characteristic of microcapsules based on various type of coating materials

Analysis	Type of coating material		
	MDE-GA	MDE-GA- β CD	MDE- β CD
<i>L</i> *	70.86 \pm 0.80 ^b	70.70 \pm 0.02 ^b	68.45 \pm 0.49 ^a
<i>Hue</i> (°)	87.38 \pm 0.77 ^a	87.26 \pm 0.68 ^a	87.79 \pm 0.11 ^a
<i>Chroma</i>	8.81 \pm 0.43 ^a	9.41 \pm 1.34 ^a	9.83 \pm 0.95 ^a

Values are the average of the standard deviation of 5 replicates. Different superscripts in the same line show statistically significant differences ($p < 0.05$), as determined by the LSD test.

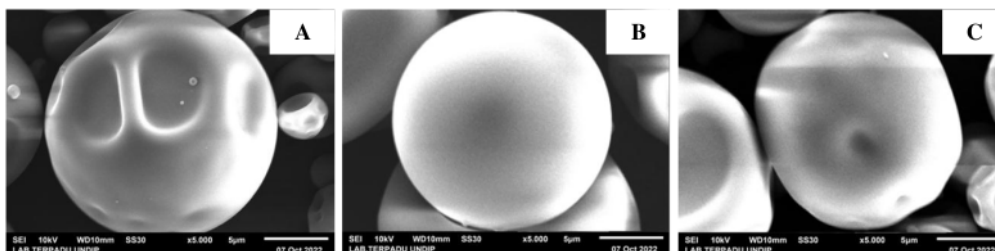


Fig (1). SEM micrograph of microcapsules with (A) MDE-AG; (B) MDE-AG- β CD; and (C) MDE- β CD

6. Morphology of Microcapsules

The morphology of microcapsules with MDE-AG, MDE-AG- β CD and MDE- β CD was shown in **Fig. (1)**. All types of coating materials produce particles in a spherical shape. The MDE-AG microcapsule has a surface that tends to be dented on several sides, tiny particles attached to the surface, and the MDE- β CD microcapsule. In comparison, MDE-AG- β CD produces microcapsules with a perfect moon shape and a smooth surface. The inclusion complex phenomenon is seen in MDE-AG- β CD. The lipophilic β CD cavity provides a suitable environment for trapping umami compounds from *Sargassum aquifolium* extract; this is confirmed in the data of Table. 1, where the highest levels of microcapsule L-glutamic acid were obtained with the MDE-AG- β CD coating material. The ability of β CD to form complexes is also influenced by many factors, such as the size of the guest molecule and the thermodynamic interaction between the guest molecule and the solvent. The formation of hydrogen bonds between β CD and flavour molecules will stabilize the inclusion complex that is formed (**Saffationpour, 2019**). Flavour inclusion complexes with β CD are known to form large aggregates in water. These aggregates usually have a regular geometry and are thought to increase the thermal stability of the liquid during drying. The excellent film-forming ability of AG also promotes the protection of flavour components during drying (**Mahdi *et al.*, 2020**). This is why MDE-AG- β CD microcapsules have a smooth surface and a perfectly round shape.

7. Particle Size Distributions

Particle size distribution of microcapsules ranged from 1.119 – 98.50 μ m (**Table 3**). Not much different from the report by **Nurhidajah *et al.* (2021)**, who observed the

particle size of microcapsules (0.19 – 94.57 μm) with the same material, and was lower than the report by *Mayasari et al. (2020)*, who used MDE as a coating material for the source of umami (43.14- 101.71 μm).

Table (3): Particle size distribution parameters of seafood flavor enhancer microcapsule by different wall materials

Wall materials	$d(4,3)/\mu\text{m}$	$d(3,2)/\mu\text{m}$	$d(0.1)/\mu\text{m}$	$d(0.5)/\mu\text{m}$	$d(0.9)/\mu\text{m}$
MDE-AG	37.688	30.828	19.238	34.096	60.373
MDE-AG- β CD	43.854	2.304	1.119	10.540	98.530
MDE- β CD	26.129	9.809	4.508	16.013	56.866

The distribution of each combination of coating materials has been observed (**Fig. 2**); MDE-AG coating materials produce the largest size with an average surface area of particles reaching 30.828 μm . Particle size of MDE-AG is 3-15 times larger than that of MDE-AG- β CD (2.304 μm) and MDE- β CD (9.809 μm) coatings. AG as a coating material will increase the stability of the emulsion, as seen from the increase in the viscosity of the liquid formed (*Dickinson et al., 2009*). Previous studies explained that a higher emulsion viscosity would contribute to the larger droplets formed during atomization, resulting in larger particles (*Fernandes et al., 2014*). Particle size is an important characteristic of microcapsules because of its strong influence on the dispensability of microcapsules. Microcapsules with small particle sizes have better dispensability to water. *Nurhidajah et al. (2021)* reported that particles with small diameters require a longer wetting time due to lower particle porosity.

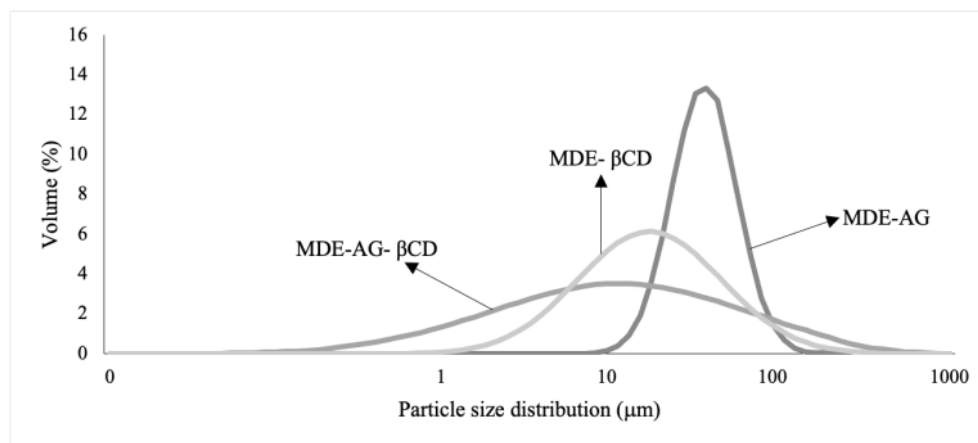


Fig (2). Particle size distribution of microcapsules by different coated materials

CONCLUSION

This study succeeded in achieving its goal of making seafood flavour enhancer microcapsules from free amino acid extracts of *Sargassum aquifolium* using MDE, AG and β CD coating materials. The addition of AG and β CD in the coating material formula significantly increased the yield and levels of microcapsule L-glutamic acid. Not only

that, improvements were also seen in the quality of microcapsules ¹ in the form of moisture content, water activity, and product hygroscopicity. Although the solubility and colour attributes tend to decrease, the resulting microcapsules have a smaller size with a smooth and perfectly round surface shape. Therefore the use of MDE combined with AG and β CD as a coating material is highly recommended in the manufacture of seafood flavour enhancer microcapsules.

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