Malaysian Journal of Chemical Engineering & Technology 4 (2) (2021) 114–123

Malaysian Journal of Chemical Engineering & Technology

e-ISSN: 2682-8588

Journal Homepage: http://myjms.mohe.gov.my/index.php/mjcet

Essential oils encapsulation performance evaluation: A review on encapsulation parameters

Siti Afiqah 'Aisyah Murtadza, Junaidah Jai *, Nurul Asyikin Md Zaki, Fazlena Hamzah

School of Chemical Engineering, College of Engineering, Universiti Teknologi MARA, Selangor, Malaysia

*Corresponding email: junejai@uitm.edu.my

Abstract

Application of essential oils (EOs) in food preservation and products is not a virgin trend owing to its bio-functional properties such as antioxidants, antimicrobials, medicinal values, and aromatic functionalities. However, EOs are prone to degrade upon exposure to different environmental surroundings, eventually losing their bio-functional activities and limits their potential applications. Hence, encapsulation process is introduced to overcome this issue. In order, to evaluate encapsulation process, there are several key indicators, known as encapsulation parameters, that reflects the performance of encapsulation process and quality of encapsulation products (encapsulates) namely encapsulation efficiency, encapsulation yield, payload/loading capacity, and surface loading. Since some terms are used interchangeably across literatures, problems arise when it comes to compare these parameters among published works as there is no specific guideline or specific term to classify these parameters. Therefore, this paper aims to help researchers understand an insight of the definition of encapsulation parameters used in evaluating performance of encapsulation process and encapsulation products of EOs. Commonly used evaluation techniques as well as some recommendations for considerations are also highlighted. Different calculation formulae used in evaluating encapsulation performance would have significant difference to the encapsulation parameters values.

Article Info

https://doi.org/10.24191/mjcet.v4i2.14481

Article history:

Received date: 29 July 2021
Accepted date: 14 October 2021
Published date: 31 October 2021

Keywords:

Encapsulation Efficiency
Encapsulation Yield
Microencapsulation
Controlled Release
Essential Oils

1.0 Introduction

Essential oils (EOs) can be extracted from any part of plants and are considered as secondary metabolites. They are usually comprised of a complex mixture of compounds such as alkaloids, flavonoids, isoflavones, monoterpenes, phenolic acids, carotenoids, and aldehydes (Seow et al., 2014). EOs consist of wide spectrum of components in which the efficacy as antimicrobial, antioxidants etc., come from synergistic effect of many components. These components are responsible for the ability of EOs to be introduced and incorporated in many applications such as in cosmetics, nutraceuticals, and food products. Application of EOs is often limited as they are susceptible to environmental conditions such as light, oxygen, and temperature; easily evaporate, nearly insoluble in water, and have strong lipophilicity and volatility (Ju et al., 2019). As a result, exploring the potential to extend their applications has become a key research issue.

Encapsulation has been introduced to improve EOs applications. It allows for preservation of bio-functional properties of EOs; improving its stability against harsh conditions, gives benevolent masking effect as well as provides controlled release of EOs. In a study by Shetta et al. (2019), it was found that encapsulation significantly enhance the thermal stability of encapsulated peppermint and green tea EOs around 2.18 and 1.74 folds, respectively as compared to the pure EOs. Encapsulation can be achieved by many techniques and could be divided into three categories; 1) chemical method; 2) physico-mechanical method; and 3) physico-chemical method. In many applications, encapsulation process might involve more than one technique (Kavousi et al., 2018). Selection of the most feasible technique would depend on the type of coated material, the operational cost, and the application of the encapsulation products. Encapsulation parameters such as encapsulation efficiency, encapsulation yield, payload/loading capacity, and surface loading are commonly used as primary indicators to reflect the performance of encapsulation process and quality of encapsulation products (encapsulates).

2.0 Encapsulation process evaluation method

Generally in encapsulation, the idea of quantifying EO upon encapsulation process is 1) to calculate encapsulation efficiency and other encapsulation parameters; 2) to perform a controlled release study and understand the kinetics of release (Rosli et al., 2018); as well as 3) to evaluate the stability of encapsulates based on how much oil is left in the encapsulates (Chung et al., 2013), or how much oil is released to the releasing media (Cortés-Camargo et al., 2019), and still adhered to the surface (Ngamekaue & Chitprasert, 2019). Besides that, it is important to exactly determine the components that are successfully encapsulated and responsible for the bio-function of EOs. These components or type of EO would have effects on encapsulation evaluation parameters. In a study by (Ardiansyah Rukmana et al., 2017), different encapsulation efficiency values were obtained when encapsulating kaffir lime oil from peels (KLO-P) and twigs oil fraction (KLO-TF) using chitosan as wall material. It was found that the encapsulation efficiency of KLO-TF is greater than KLO-P. The encapsulation efficiency difference was claimed to be attributed by the components presented in each kaffir lime oil in which KLO-TF contains more oxygenated monoterpene components while KLO-P is dominated by the hydrocarbon monoterpenes components. Oxygenated monoterpenes components are more likely to interact with the functional group (active site) in the encapsulate and as a result, more KLO-TF was successfully encapsulated.

Determination of EO in encapsulates can be done gravimetrically through direct measuring (Hsieh et al., 2006; Siow & Ong, 2012) or distillation process. However, drawbacks associate with such techniques

Table 1: Method and solvent used for digestion of encapsulates

Digestion method	Type of EOs	Wall materials	Solvent used	References
Physical	Citronella EOs	Gelatine/Sodium sulphate	Chloroform	(Solomon et al., 2012)
	Zanthoxylum limonella Oil	Chitosan/Gelatine	Tween 80	(Maji et al., 2007)
	Neem seed oil	Gelatine A/Sodium Carboxymethyl cellulose	Tween 80	(Devi & Maji, 2011)
	Kaffir lime oil	Chitosan	n-hexane	(Ardiansyah Rukmana et al., 2017)
	Citronella oil	Leather waste gelatine/ Sodium alginate	Hexane	(De Matos et al., 2018)
	Palm oil	Chitosan/ Xanthan and chitosan/pectin	Hexane	(Rutz et al., 2017)
	Lemon EOs	Chitosan/hicap	Hexane	(Hasani et al., 2018)
	Cinnamaldehyde	Gelatine/Pectin	Hexane	(Muhoza et al., 2019)
	Peppermint oil	Chitosan/Alginate	Ethanol	(Deka et al., 2016)
	Thyme oil	Melamine–formaldehyde	Methanol	(Chung et al., 2013)
	Holy basil essential oil	Gelatine/ beeswax-carboxymethyl cellulose composite	Dichloromethane	(Ngamekaue & Chitprasert, 2019)
	Kaffir lime oil	Gum Arabic/Maltodextrin	-	(Triyono et al., 2018)
Chemical	Clove/Cinnamon/Thyme oil	Alginate	n-hexane	(Soliman et al., 2013)
	Moxa oil	Gelatine/Gum Arabic	Hexane	(Li et al., 2013)
	Tuna oil	Gelatine/sodium hexametaphosphate	Hexane	(Wang et al., 2014)
	Krill oil	Krill protein isolated with isoelectric solubilisation/precipitation (ISP)	Petroleum ether	(Shi et al., 2018)
Enzymatic	Vetiver EOs	Gelatine-Gum Arabic	Dichloromethane	(Prata et al., 2008)
Thermal	Kaffir lime oil	Konjac glucomannan (KGM) and Gum Arabic (GA).	-	(Borompichaichartkul et al., 2012)
	Lemon EOs	Mesquite gum/ chia mucilage	-	(Cortés-Camargo et al., 2019)
	Sweet orange oil	Soybean protein isolate/ Gum Arabic	-	(Jun-xia et al., 2011)
Physical-thermal	D-limonene	*Alyssum homolocarpum* seed gum	Hexane	(Khoshakhlagh et al., 2017)
Chemical-Physical	Fish oil	Gelatine/Gum-Arabic	-	(Yu et al., 2017)

are that large amount of formulation is required, improper extraction, and chances of loss of EO due to volatilisation (Vishwakarma et al., 2016). To overcome these issues, reliable techniques using analytical methods such as chromatographic or spectrophotometric methods are introduced and expected to exhibit higher values than when thermogravimetric analysis is used (Xiao et al., 2014). When employing these analytical methods, sometimes, digestion of the wall material is required in which it can be achieved physically, chemically, or enzymatically (Wang et al., 2018). Table 1 below shows different types of EOs and commonly used solvents and methods to digest encapsulates wall. Subsequently, EO is extracted using organic solvent such as hexane (Khoshakhlagh et al., 2017), petroleum ether (Zhang et al., 2012), ethanol (Tolun et al., 2016) or non-ionic surfactant; tween-80 (Devi & Maji, 2011) before proceeded for quantification using appropriate analytical methods. These analytical methods also have some disadvantages such as possible experimental error, chances of loss of EO due to volatilisation and possibilities that method selected is not convenient. For example, in cases where digestion of encapsulates walls is needed, the digested wall materials might somehow interfere with the spectrometric reading of EO. However, this could be resolved by using appropriate solvent and technique. Li et al. (2013) used hexane to extract Moxa oil from encapsulates since gelatine and Gum Arabic that were used as encapsulating material did not interfere with the measurement process as they were insoluble in hexane. Meanwhile, Fraj et al. (2021) used derivative spectrophotometry for quantitative analysis of core material since wall materials used (vitamin C and genipin) were also soluble in ethanol.

3.0 Encapsulation evaluation parameters

In order, to evaluate encapsulation process performance, several parameters that are commonly used by many researchers are encapsulation efficiency, encapsulation yield, payload/ loading capacity and surface loading. These parameters are greatly affected by factors such as ratio of wall materials to EO used, type of crosslinker and encapsulation techniques used to encapsulate. Ideally, encapsulation parameters are to be obtained as high value as possible (Wang et al., 2018). Currently, improvement on the encapsulation parameters and controlled release is achieved by double encapsulation technique in which EO is coated with second layer wall (Yu et al., 2017). However, these parameters involve calculations that slightly vary in different publications. Researchers might tend to overlook the effect of different calculations used and simply make comparison and draw conclusions on the efficiency of their encapsulation process without addressing the errors that might occur from different calculations used in evaluating encapsulation parameters.

3.1 Encapsulation efficiency (EE%)

Encapsulation efficiency (EE) is defined as the percentage of EO that is successfully entrapped within the wall material over the EO introduced in the beginning of the process (De Matos et al., 2018). This term is widely used in many studies and publications to reflect the performance of encapsulation process. Other terms such as entrapment efficiency (Dubey et al., 2020; Merodio et al., 2001), internalizing efficiency (Rutz et al., 2017) and loading efficiency (Chung et al., 2013) are also used to describe the encapsulation performance process. Table 2 summarises EE formulae that are used widely in many literatures. In some studies Eq. (2) is often considered as encapsulation yield (e.g., Bakry et al., 2019; Jun-xia et al., 2011; Muhoza et al., 2019).

Determination of EE value would require consideration as whether to include surface EO as part of encapsulated EO or not. In cases where surface EO is not considered as part of encapsulated EO, Eq. (1), Eq. (3), Eq. (4), and Eq. (5) are often used. Calculation of EE using Eq. (3), Eq. (4), and Eq. (5) would require determination of surface EO value. Usually, this surface EO is firstly removed using appropriate solvent and quantified. Other information required would include total EO used to prepare the same amount of encapsulates and information on amount of EO in certain amount of encapsulates before surface EO removal. According to Carneiro et al. (2013), total EO was assumed to be equal to the initial EO added to the system, since preliminary tests revealed that all initial EO used in the experiment was retained since EO is not volatile

In Eq. (2), surface EO is not removed and is included in calculating the EE value. This is because, at the early stage when encapsulates are collected prior to the encapsulation process, surface EO could be considered as part of EO that is successfully entrapped within encapsulates. This is common especially in encapsulation which involves thermal treatment where

Table 2: Encapsulation efficiency (EE%) formulae widely used in literatures

Calculation formulae		Description	References
$\mathbf{EE\% = \frac{W_C}{W_O} \times 100\%}$	Eq. (1)	W_C is weight (g) of the EO in certain amount of encapsulates after surface EO removal W_O is weight (g) of the EO used to prepare the same amount of encapsulates.	(Devi & Maji, 2011) (Chung et al., 2013)
$\mathbf{EE\% = \frac{W_T}{W_O} \times 100\%}$	Eq. (2)	W_T is weight (g) of the EO in certain amount of encapsulates before surface EO removal W_O is weight (g) of the EO used to prepare the same amount of encapsulates.	(Rungwasantisuk & Raibhu, 2020)
$\mathbf{EE\% = \frac{W_O - W_S}{W_O} \times 100\%}$	Eq. (3)	W_S is weight (g) of the surface EO in certain amount of encapsulates W_O is weight (g) of the EO used to prepare the same amount of encapsulates	(Girardi et al., 2017)
$\mathbf{EE\% = \frac{W_T - W_S}{W_T} \times 100\%}$	Eq. (4)	W_T is weight (g) of the EO in certain amount of encapsulates before surface EO removal W_S is weight (g) of the surface EO in certain amount of encapsulates	(Wang et al., 2014)
$\mathbf{EE\% = \frac{W_T - W_S}{W_O} \times 100\%}$	Eq. (5)	W_T is weight (g) of the EO in certain amount of encapsulates before surface EO removal W_S is weight (g) of the surface EO in certain amount of encapsulates W_O is weight (g) of the EO used to prepare the same amount of encapsulates	(Rutz et al., 2017) (Muhoza et al., 2019) (Yu et al., 2017)
$\mathbf{OT\% = \frac{WI_O}{WI_C} \times 100\%}$	Eq. (6)	OT% is the theoretical EO loading WIo is the initial mass of EO added to the system WIc is the initial mass of the encapsulates	(Bastos et al., 2020) (da Silva Soares et al., 2019)
$\mathbf{OC\% = \frac{WF_O}{WF_C} \times 100\%}$	Eq. (7)	OC% is the actual EO loading WFo is the actual EO content after encapsulation	
$\mathbf{EE\% = \frac{OC\%}{OT\%} \times 100\%}$	Eq. (8)	WFc is the final mass of encapsulates after encapsulation process EE% is the percentage of the actual EO loading divided by the percentage of the theoretical EO loading	

volatilised EO might move outwards and eventually deposited on the encapsulates surface. However, in a freshly prepared encapsulates, the amount of this surface EO is relatively less compared to encapsulated EO (Ngamekaue & Chitprasert, 2019).

Nevertheless, in either way (removal or non-removal surface EO), the higher the value of EE indicates a more efficient process. An efficient process means less EO is adsorbed on the surface of the encapsulates and/or not encapsulated in the process. EO adsorbed on the surface are often unfavourable as they could promote the release of EO from encapsulates leading to a higher oxidation and/or degradation of encapsulates (Wang et al., 2018). According to Zhang et al. (2012), initial burst release observed in the kinetic study of microalgal oil release from encapsulates was claimed to be caused by distribution of this EO near the surface of the encapsulates. In a study by Rutz et al. (2017), Eq. (4) and Eq. (5) are used to evaluate the effect of wall materials and drying methods on the encapsulation process. Results obtained calculated using both formulae have quite a significant difference. The same findings are also found in a study by Shi et al. (2018). Thus, in evaluating encapsulation process, one should carefully define the efficiency term used in their experiments and how they calculate their EE when they try to make comparison with other findings. One should also decide whether surface EO should be included in the calculation or not as it will affect the EE value. Theoretically, calculation which includes surface EO as a part of encapsulated EO would result in a relatively higher encapsulation efficiency value as can be seen in comparison Table 3 between EE value calculated using Eq. (1) and Eq. (2).

3.2 Surface loading (SL%)

Surface loading (SL) is defined as the percentage of weight of EO deposited on the surface of the certain amount of encapsulates over the weight of certain amount of encapsulates. It can also represent the amount of unencapsulated EO in the process. It can be calculated using Eq. (9) (Ngamekaue & Chitprasert, 2019) and Eq. (10) (Khoshakhlagh et al., 2017) as in Table 4.

In most cases and applications, it is desirable for EO to be entrapped inside the protection layer (wall) at the end of the encapsulation process. However, according to Wang et al. (2018), normally some EOs are more likely to be adsorbed on the outer surface of the encapsulates rather than entrapped within the wall material. Surface EO is regarded as free EO that surrounds and exists at the external layer of the encapsulates and can be extracted commonly by using organic solvent (Tirgar et al., 2015). Selection of suitable organic solvent that could dissolve the surface EO is important as to ensure that the solvent does not disrupt the encapsulates (Muhoza et al., 2019). For example, Nori et al. (2011) added 2.0 mL of ethanol to 0.2 g of sample so that the alcohol could dissolve the propolis that was outside the encapsulates, without disruption. Force is often introduced during extraction not to rupture the wall but to enhance the extraction of surface EO. The solvent mixture is then filtered to separate between encapsulates and the solvent which contains surface EO. Extracted surface EO is then subjected for determination which could be done gravimetrically or by using any appropriate analytical instruments. Over time, encapsulated EO might be released from encapsulates by different release mechanisms such as diffusion, polymer relaxation (swelling/shrinking), erosion, or fragmentation

Table 3: Comparison between EE (%) value calculated using Eq. (1) and Eq. (2)

	EE (%) values	**References**
EE calculated using Eq. (1)	2.00–3.67 3.27–14.29	(Ardiansyah Rukmana et al., 2017)
	22.6–77.5	(Chung et al., 2013)
	32–60	(Maji & Hussain, 2008)
	36.2–62.8	(Solomon et al., 2012)
	74.76–95.23	(Devi & Maji, 2011)
	90–94	(Soliman et al., 2013)
EE calculated using Eq. (2)	66–98 62–98	(Siow & Ong, 2012)
	73.7	(De Matos et al., 2018)
	76.1–84.7	(Rungwasantisuk & Raibhu, 2020)
	87–93	(Khoshakhlagh et al., 2017)
	93.71	(Yu et al., 2017)

Table 4: Surface Loading (SL%) formulae used in literatures

``		**Description**	**References**
$\mathbf{SL\%} = \frac{\mathbf{W_S}}{\mathbf{W_W}} \times \mathbf{100\%}$	Eq. (9)	W_S is weight (g) of the surface EO in certain amount of encapsulates W_W is weight (g) of the certain amount of encapsulates before surface EO removal	(Ngamekaue & Chitprasert, 2019) (Bakry et al., 2019)
$\mathbf{SL\%} = \frac{\mathbf{W_S}}{\mathbf{W_T}} \times \mathbf{100\%}$	Eq. (10)	W_S is weight (g) of the surface EO in certain amount of encapsulates W_T is weight (g) of the EO in certain amount of encapsulates before surface EO removal	(Khoshakhlagh et al., 2017)

(Vishwakarma et al., 2016). It is possible that this previously encapsulated EO escaped from encapsulates but still adhered on the surface of the encapsulates. Quantification of this surface EO could be used to evaluate the controlled release of EO from encapsulates or represent the stability of encapsulates (Ngamekaue & Chitprasert, 2019).

3.3 Encapsulation yield (EY%)

Encapsulation yield (EY) is defined as the percentage of encapsulates produced over the total weight materials used in the process (Khoshakhlagh et al., 2017). It can be calculated using Eq. (11) as in Table 5. Depending on encapsulates preparation, some studies would include mass of crosslinker (Devi & Maji, 2011; Hussain et al., 2013) and EO (core material) (Rutz et al., 2017) in the calculation of total formulations used to prepare the encapsulates. In many processes, high EY value is desired as it means that high encapsulates can be produced over introduced encapsulation materials. Identifying EY allows us to estimate how much encapsulates to produce or would produce in an encapsulation process. However, relying on the EY to evaluate encapsulation process is not convincing enough. It is still arguable as though the encapsulates are successfully obtained, one could still wonder if the EO is really entrapped within the encapsulates or is it just emptied encapsulates. Though qualitative analysis could resolve this issue by examining the structural or physical characteristics of the encapsulates using appropriate microscopic instruments, another possible method is by calculating the payload or loading capacity. Determining this parameter is important especially in real industrial applications where unsuccessful encapsulation process is not a favourable option.

3.4 Payload/loading capacity (LC%)

Payload or loading capacity (LC) represents the percentage of EO that is presented in certain amount of encapsulates. The formulae used to calculate the LC is represented by Eq. (12), Eq. (13) and Eq. (14) in Table 6. In the calculation of LC, some study would include surface EO (e.g., Deka et al., 2016) in their calculations. Meanwhile, there are studies that will firstly remove surface EO before extracting encapsulated EO and proceed with LC calculation (e.g., Devi & Maji, 2011). Loading capacity provides information on how much of the EO that could be loaded within encapsulates. The higher the value of LC indicates higher percentage of EO presents in certain amount of encapsulates. It is often achieved with increasing EO to wall ratio concentration (Deka et al., 2016). When higher LC is achieved, it means that less polymers/wall materials are needed to perform encapsulation (Shi et al., 2018). This information is crucial in designing a feasible encapsulation process of lower operational cost. Higher LC leads to a faster

Table 5: Encapsulation yield (EY%) formulae used in literatures

Calculation formulae		Description	References
$\mathbf{EY\% = \frac{W_W}{W_{all}} \times 100\%}$	Eq. (11)	W_W is weight (g) of the certain amount of encapsulates before surface EO removal. W_{all} is weight (g) of the total formulations used to prepare the encapsulates (wall material, EO and crosslink; if any).	(Khoshakhlagh et al., 2017) (Rutz et al., 2017)

Table 6: Loading capacity (LC%) formulae used in literatures

Calculation formulae		Description	References
$\mathbf{LC\% = \frac{W_C}{W_{WR}} \times 100\%}$	Eq. (12)	W_C is weight (g) of the EO in certain amount of encapsulates after surface EO removal. W_{WR} is weight (g) of the certain amount of encapsulates after surface EO removal.	(Devi & Maji, 2011)
$\mathbf{LC\% = \frac{W_T}{W_W} \times 100\%}$	Eq. (13)	W_T is weight (g) of the EO in certain amount of encapsulates before surface EO removal W_W is weight (g) of the certain amount of encapsulates before surface EO removal.	(Deka et al., 2016) (Shi et al., 2018) (Wang et al., 2014)
$\mathbf{LC\% = \frac{W_C}{W_W} \times 100\%}$	Eq. (14)	W_C is weight (g) of the EO in certain amount of encapsulates after surface EO removal. W_W is weight (g) of the certain amount of encapsulates before surface EO removal.	(Khoshakhlagh et al., 2017) (Yu et al., 2017)

release rate of encapsulated EO due to enhancement of driving force (Khoshakhlagh et al., 2017). Besides that, the increase in LC is not in proportion with encapsulation efficiency. At certain saturation limit, LC will start to decrease. Saturation limit is where the external phase forming wall materials is unable to tolerate any addition of EO and eventually losing its emulsifying capacity (Deka et al., 2016; Shi et al., 2018). Hence, more EO is unencapsulated and/or adhered to the surface of encapsulates rather than entrapped within encapsulates. Determination of the suitable proportion between EO and wall material concentration is crucial to achieve efficient encapsulation process. Another significance of identifying LC is to proceed with controlled release and kinetic study of encapsulates. LC gives information on the initial amount of EO entrapped and how long the encapsulates can hold the active ingredient in given time (Rosli et al., 2018).

3.5 Moisture content

According to Yuen et al. (2012), weight of encapsulates is affected by the moisture content if encapsulates obtained and used in the calculation of encapsulation parameters is in wet form. This is because, different batch of encapsulates produced would have different moisture content, thus, make it difficult to compare between different batches. For example, encapsulates produced from coacervation process with no subsequent drying method. Moisture content rate (MCR%) is introduced (Dong et al., 2011; Dong et al., 2007; Yuen et al., 2012) to give accurate value of encapsulation parameters which involve the weight of encapsulates in the calculation formula. The formulae for the calculation of moisture content rate and weight of pseudo dry encapsulates (W_{dry}) is written as in Table 7. Calculation for weight of pseudo dry encapsulates (W_{dry}); Eq. (17):

$$W_{dry} = W_{WT} \times (1 - MCR) \qquad (17)$$

where: MCR is moisture content rate

Therefore, to obtain a more accurate value of encapsulation parameters for encapsulates obtained in a wet form, the weight of pseudo dry encapsulates (W_{dry}) is used in the calculation of encapsulation parameters.

4.0 Conclusions

Several important encapsulation parameters including encapsulation efficiency, encapsulation yield, payload/ loading capacity, surface loading, evaluation techniques and few recommendations to be considered while evaluating these parameters have been discussed. Encapsulation technology has always been a promising area to be explored owing to its ability of providing a controlled release. Therefore, future studies on designing encapsulation system using new materials that are sustainable should be conducted to extend its potential integration into many areas of applications in industry.

Table 7: Moisture content rate (MCR%) formulae used in literatures

Calculation formulae		Description	References
$\mathbf{MCR\%} = \frac{(\mathbf{W_{WT}} - \mathbf{W_{wt}}) - (\mathbf{W_E} - \mathbf{W_e})}{\mathbf{W_{WT}}} \times \mathbf{100\%}$	Eq. (15)	W_{WT} is weight (g) of the certain amount of wet encapsulates before total dryness W_{wt} is weight (g) of the certain amount of wet encapsulates after total dryness $(W_{WT} - W_{wt})$ is total mass of released oil and water, W_E is weight (g) of the EO in certain amount of wet encapsulates before total dryness W_e is weight (g) of the EO in certain amount of wet encapsulates after total dryness $(W_E - W_e)$ is mass of released oil	(Dong et al., 2007) (Dong et al., 2011)
$\mathbf{MCR\%} = \frac{(\mathbf{W_{WT}} - \mathbf{W_{wt}})}{\mathbf{W_{WT}}} \times \mathbf{100\%}$	Eq. (16)	W_{WT} is weight (g) of the certain amount of wet encapsulates before total dryness W_{wt} is weight (g) of the certain amount of wet encapsulates after total dryness	(Yuen et al., 2012)

Acknowledgement

This work was funded by Malaysia's Ministry of Education (MOE) Fundamental Research Grant Scheme (FRGS) through 600-RMI/FRGS/5/3 (186/2019) grant. We are very grateful for the assistance provided by all parties in assisting the research activities.

References

Ardiansyah Rukmana, R. A. F., Warsito, W., & Rahman, M. F. (2017). Microencapsulation of Kaffir Lime Oil (citrus hystrix d. c.) from Peels and Twigs Oil Fraction and its Activity as Antioxidant. *Proceedings of International Conference of Essential Oil (ICEO 2017)*, 112–118.

Bakry, A. M., Huang, J., Zhai, Y., & Huang, Q. (2019). Myofibrillar protein with κ- or λ-carrageenans as novel shell materials for microencapsulation of tuna oil through complex coacervation. *Food Hydrocolloids*, *96*, 43–53. https://doi.org/10.1016/j.foodhyd.2019.04.070

Borompichaichartkul, C., Srzednicki, G., Piriyapunsakul, S., Panket, W., Zhao, J., & Adamiec, J. (2012). Microencapsulation of kaffir lime oil and its functional properties. *Drying Technology*, *30*(9), 914–920. https://doi.org/10.1080/07373937.2012.666777

Bastos, L. P. H., dos Santos, C. H. C., de Carvalho, M. G., & Garcia-Rojas, E. E. (2020). Encapsulation of the black pepper (*Piper nigrum* L.) essential oil by lactoferrin-sodium alginate complex coacervates: Structural characterization and simulated gastrointestinal conditions. *Food Chemistry*, *316*(January), 126345. https://doi.org/10.1016/j.foodchem.2020.126345

Carneiro, H. C. F., Tonon, R. V., Grosso, C. R. F., & Hubinger, M. D. (2013). Encapsulation efficiency and oxidative stability of flaxseed oil microencapsulated by spray drying using different combinations of wall materials. *Journal of Food Engineering*, *115*(4), 443–451. https://doi.org/10.1016/j.jfoodeng.2012.03.033

Chung, S. K., Seo, J. Y., Lim, J. H., Park, H. H., Yea, M. J., & Park, H. J. (2013). Microencapsulation of essential oil for insect repellent in food packaging system. *Journal of Food Science*, *78*(5), 1–6. https://doi.org/10.1111/1750-3841.12111

Cortés-Camargo, S., Acuña-Avila, P. E., Rodríguez-Huezo, M. E., Román-Guerrero, A., Varela-Guerrero, V., & Pérez-Alonso, C. (2019). Effect of chia mucilage addition on oxidation and release kinetics of lemon essential oil microencapsulated using mesquite gum – Chia mucilage mixtures. *Food Research International*, *116*, 1010–1019. https://doi.org/10.1016/j.foodres.2018.09.040

da Silva Soares, B., Siqueira, R. P., de Carvalho, M. G., Vicente, J., & Garcia-Rojas, E. E. (2019). Microencapsulation of *sacha inchi* oil (*Plukenetia volubilis* L.) using complex coacervation: Formation and structural characterization. *Food Chemistry*, *298*(February), 125045. https://doi.org/10.1016/j.foodchem.2019.125045

De Matos, E. F., Scopel, B. S., & Dettmer, A. (2018). Citronella essential oil microencapsulation by complex coacervation with leather waste gelatin and sodium alginate. *Journal of Environmental Chemical Engineering*, *6*(2), 1989–1994. https://doi.org/10.1016/j.jece.2018.03.002

Deka, C., Deka, D., Bora, M. M., Jha, D. K., & Kakati, D. K. (2016). Synthesis of peppermint oil-loaded chitosan/alginate polyelectrolyte complexes and study of their antibacterial activity. *Journal of Drug Delivery Science and Technology*, *35*, 314–322. https://doi.org/10.1016/j.jddst.2016.08.007

Devi, N., & Maji, T. K. (2011). Study of complex coacervation of gelatin a with sodium carboxymethyl cellulose: Microencapsulation of neem (*Azadirachta Indica* A. Juss.) seed oil (NSO). *International Journal of Polymeric Materials and Polymeric Biomaterials*, *60*(13), 1091–1105. https://doi.org/10.1080/00914037.2011.553851

Dong, Z. J., Touré, A., Jia, C. S., Zhang, X. M., & Xu, S. Y. (2007). Effect of processing parameters on the formation of spherical multinuclear microcapsules encapsulating peppermint oil by coacervation. *Journal of Microencapsulation*, *24*(7), 634–646. https://doi.org/10.1080/02652040701500632

Dong, Z., Ma, Y., Hayat, K., Jia, C., Xia, S., & Zhang, X. (2011). Morphology and release profile of microcapsules encapsulating peppermint oil by complex coacervation. *Journal of Food Engineering*, *104*(3), 455–460. https://doi.org/10.1016/j.jfoodeng.2011.01.011

Dubey, V., Mohan, P., Dangi, J. S., & Kesavan, K. (2020). Brinzolamide loaded chitosan-pectin mucoadhesive nanocapsules for management of glaucoma: Formulation, characterization and pharmacodynamic study. *International Journal of Biological Macromolecules*, *152*, 1224–1232. https://doi.org/10.1016/j.ijbiomac.2019.10.219

Fraj, J., Petrović, L., Đekić, L., Budinčić, J. M., Bučko, S., & Katona, J. (2021). Encapsulation and release of vitamin C in double W/O/W emulsions followed by complex coacervation in gelatin-sodium caseinate system. *Journal of Food Engineering*, *292*(May 2020), 110353. https://doi.org/10.1016/j.jfoodeng.2020.110353

Girardi, N. S., García, D., Passone, M. A., Nesci, A., & Etcheverry, M. (2017). Microencapsulation of *Lippia turbinata* essential oil and its impact on peanut seed quality preservation. *International Biodeterioration and Biodegradation*, *116*, 227–233. https://doi.org/10.1016/j.ibiod.2016.11.003

Hasani, S., Ojagh, S. M., & Ghorbani, M. (2018). Nanoencapsulation of lemon essential oil in Chitosan-Hicap system. Part 1: Study on its physical and structural characteristics. *International Journal of Biological Macromolecules*, *115*, 143–151. https://doi.org/10.1016/j.ijbiomac.2018.04.038

Hsieh, W. C., Chang, C. P., & Gao, Y. L. (2006). Controlled release properties of Chitosan encapsulated volatile citronella oil microcapsules by thermal treatments. *Colloids and Surfaces B: Biointerfaces*, *53*(2), 209–214. https://doi.org/10.1016/j.colsurfb.2006.09.008

Hussain, M. R., Iman, M., & Maji, T. K. (2013). Determination of degree of deacetylation of chitosan and their effect on the release behavior of essential oil from chitosan and chitosan-gelatin complex microcapsules. *International Journal of Advanced Engineering Applications*, *6*(4), 4–12. https://doi.org/10.1152/jn.00961.2010

Ju, J., Chen, X., Xie, Y., Yu, H., Guo, Y., Cheng, Y., Qian, H., & Yao, W. (2019). Application of essential oil as a sustained release preparation in food packaging. *Trends in Food Science and Technology*, *92*(1800), 22–32. https://doi.org/10.1016/j.tifs.2019.08.005

Jun-xia, X., Hai-yan, Y., & Jian, Y. (2011). Microencapsulation of sweet orange oil by complex coacervation with soybean protein isolate/gum Arabic. *Food Chemistry*, *125*(4), 1267–1272. https://doi.org/10.1016/j.foodchem.2010.10.063

Kavousi, H. R., Fathi, M., & Goli, S. A. H. (2018). Novel cress seed mucilage and sodium caseinate microparticles for encapsulation of curcumin: An approach for controlled release. *Food and Bioproducts Processing*, *110*, 126–135. https://doi.org/10.1016/j.fbp.2018.05.004

Khoshakhlagh, K., Koocheki, A., Mohebbi, M., & Allafchian, A. (2017). Development and characterization of electrosprayed *Alyssum homolocarpum* seed gum nanoparticles for encapsulation of D-limonene. *Journal of Colloid and Interface Science (490)*. 562–575. https://doi.org/10.1016/j.jcis.2016.11.067

Li, L., Au, W., Hua, T., Zhao, D., & Wong, K. (2013). Improvement in antibacterial activity of moxa oil containing gelatin-Arabic gum microcapsules. *Textile Research Journal*, *83*(12), 1236–1241. https://doi.org/10.1177/0040517512467059

Maji, T. K., Baruah, I., Dube, S., & Hussain, M. R. (2007). Microencapsulation of *Zanthoxylum limonella* oil (ZLO) in glutaraldehyde crosslinked gelatin for mosquito repellent application. *Bioresource Technology*, *98*(4), 840–844. https://doi.org/10.1016/j.biortech.2006.03.005

Maji, T. K., & Hussain, M. R. (2009). Microencapsulation of Zanthoxylum limonella oil (ZLO) in genipin crosslinked chitosan-gelatin complex for mosquito repellent application. *Journal of Applied Polymer Science*, *111*, 779–785. https://doi.org/10.1002/app.29001

Merodio, M., Arnedo, A., Renedo, M. J., & Irache, J. M. (2001). Ganciclovir-loaded albumin nanoparticles: Characterization and in vitro release properties. *European Journal of Pharmaceutical Sciences*, *12*(3), 251–259. https://doi.org/10.1016/S0928-0987(00)00169-X

Muhoza, B., Xia, S., Cai, J., Zhang, X., Duhoranimana, E., & Su, J. (2019). Gelatin and pectin complex coacervates as carriers for cinnamaldehyde: Effect of pectin esterification degree on coacervate formation and enhanced thermal stability. *Food Hydrocolloids*, *87*, 712–722. https://doi.org/10.1016/j.foodhyd.2018.08.051

Ngamekaue, N., & Chitprasert, P. (2019). Effects of beeswax-carboxymethyl cellulose composite coating on shelf-life stability and intestinal delivery of holy basil essential oil-loaded gelatin microcapsules. *International Journal of Biological Macromolecules*, *135*, 1088–1097. https://doi.org/10.1016/j.ijbiomac.2019.06.002

Nori, M. P., Favaro-Trindade, C. S., Matias de Alencar, S., Thomazini, M., de Camargo Balieiro, J. C., & Contreras Castillo, C. J. (2011). Microencapsulation of propolis extract by complex coacervation. *LWT - Food Science and Technology*, *44*(2), 429–435. https://doi.org/10.1016/j.lwt.2010.09.010

Prata, A. S., Zanin, M. H. A., Ré, M. I., & Grosso, C. R. F. (2008). Release properties of chemical and enzymatic crosslinked gelatin-gum Arabic microparticles containing a fluorescent probe plus vetiver essential oil. *Colloids and Surfaces B: Biointerfaces*, *67*(2), 171–178. https://doi.org/10.1016/j.colsurfb.2008.08.014

Rosli, N. A., Hasham, R., & Aziz, A. A. (2018). Design and physicochemical evaluation of lipid encapsulated *Zingiber zerumbet* oil by d-optimal mixture design. *Jurnal Teknologi*, *80*(3), 105–113. https://doi.org/10.11113/jt.v80.11268

Rungwasantisuk, A., & Raibhu, S. (2020). Application of encapsulating lavender essential oil in gelatin/gum Arabic complex coacervate and varnish screen-printing in making fragrant gift-wrapping paper. *Progress in Organic Coatings*, *149*, 105924. https://doi.org/10.1016/j.porgcoat.2020.105924

Rutz, J. K., Borges, C. D., Zambiazi, R. C., Crizel-Cardozo, M. M., Kuck, L. S., & Noreña, C. P. Z. (2017). Microencapsulation of palm oil by complex coacervation for application in food systems. *Food Chemistry*, *220*, 59–66. https://doi.org/10.1016/j.foodchem.2016.09.194

Seow, Y. X., Yeo, C. R., Chung, H. L., & Yuk, H. G. (2014). Plant essential oils as active antimicrobial agents. *Critical Reviews in Food Science and Nutrition*, *54*(5), 625–644. https://doi.org/10.1080/10408398.2011.599504

Shetta, A., Kegere, J., & Mamdouh, W. (2019). Comparative study of encapsulated peppermint and green tea essential oils in chitosan nanoparticles: Encapsulation, thermal stability, in-vitro release, antioxidant, and antibacterial activities. *International Journal of Biological Macromolecules*, *126*, 731–742. https://doi.org/10.1016/j.ijbiomac.2018.12.161

Shi, L., Beamer, S. K., Yang, H., & Jaczynski, J. (2018). Micro-emulsification/encapsulation of krill oil by complex coacervation with krill protein isolated using isoelectric solubilization/precipitation. *Food Chemistry*, *244*(March 2017), 284–291. https://doi.org/10.1016/j.foodchem.2017.10.050

Siow, L., & Ong, C. (2012). Effect of pH on garlic oil encapsulation by complex coacervation. *Journal of Food Processing & Technology*, *04*(01), 1–5. https://doi.org/10.4172/2157-7110.1000199

Soliman, E. A., El-Moghazy, A. Y., El-Din, M. S. M., & Massoud, M. A. (2013). Microencapsulation of essential oils within alginate: Formulation and in vitro evaluation of antifungal activity. *Journal of Encapsulation and Adsorption Sciences*, *03*(01), 48–55. https://doi.org/10.4236/jeas.2013.31006

Solomon, B., Sahle, F. F., Gebre-Mariam, T., Asres, K., & Neubert, R. H. H. (2012). Microencapsulation of citronella oil for mosquito-repellent application: Formulation and in vitro permeation studies. *European Journal of Pharmaceutics and Biopharmaceutics*, *80*(1), 61–66. https://doi.org/10.1016/j.ejpb.2011.08.003

Tirgar, M., Jinap, S., Zaidul, I. S. M., & Mirhosseini, H. 2015). Suitable coating material for microencapsulation of spray-dried fish oil. *Journal of Food Science and Technology*, *52*(7), 4441–4449. https://doi.org/10.1007/s13197-014-1515-3

Tolun, A., Altintas, Z., & Artik, N. (2016). Microencapsulation of grape polyphenols using maltodextrin and gum Arabic as two alternative coating materials: Development and characterization. *Journal of Biotechnology*, *239*, 23–33. https://doi.org/10.1016/j.jbiotec.2016.10.001

Triyono, K., Suhartatik, N., & Wulandari, Y. W. (2018). Nanoencapsulating of kaffir lime oil with coacervation method using arabic gum and maltodextrin as encapsulant. *International Journal of Food and Nutrition Science*, *3*(1), 43–48.

Vishwakarma, G. S., Gautam, N., Babu, J. N., Mittal, S., & Jaitak, V. (2016). Polymeric encapsulates of essential oils and their constituents: A review of preparation techniques, characterization, and sustainable release mechanisms. *Polymer reviews*, *56*(4), 668–701. https://doi.org/10.1080/15583724.2015.1123725

Xiao, Z., Liu, W., Zhu, G., Zhou, R., & Niu, Y. (2014). Production and characterization of multinuclear microcapsules encapsulating lavender oil by complex coacervation. *Flavour and Fragrance Journal*, *29*(3), 166–172. https://doi.org/10.1002/ffj.3192

Wang, B., Adhikari, B., & Barrow, C. J. (2014). Optimisation of the microencapsulation of tuna oil in gelatin-sodium hexametaphosphate using complex coacervation. *Food Chemistry*, *158*, 358–365. https://doi.org/10.1016/j.foodchem.2014.02.135

Wang, B., Akanbi, T. O., Agyei, D., Holland, B. J., & Barrow, C. J. (2018). Coacervation technique as an encapsulation and delivery tool for hydrophobic bio-functional compounds. In *Role of Materials Science in Food Bioengineering* (pp. 235-261). Academic Press.

Yu, F., Li, Z., Zhang, T., Wei, Y., Xue, Y., & Xue, C. (2017). Influence of encapsulation techniques on the structure, physical properties, and thermal stability of fish oil microcapsules by spray drying. *Journal of Food Process Engineering*, *40*(6), 1–9. https://doi.org/10.1111/jfpe.12576

Yuen, C. W. M., Yip, J., Liu, L., Cheuk, K., Kan, C. W., Cheung, H. C., & Cheng, S. Y. (2012). Chitosan microcapsules loaded with either miconazole nitrate or clotrimazole, prepared via emulsion technique. *Carbohydrate Polymers*, *89*(3), 795–801. https://doi.org/10.1016/j.carbpol.2012.04.013

Zhang, K., Zhang, H., Hu, X., Bao, S., & Huang, H. (2012). Synthesis and release studies of microalgal oil-containing microcapsules prepared by complex coacervation. *Colloids and Surfaces B: Biointerfaces*, *89*(1), 61–66. https://doi.org/10.1016/j.colsurfb.2011.08.023