Control Release of Encapsulation Citronella Oil by Complex Coacervation in Simulated Clean Floor Solution

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Abstract

Citronella oil is one of the essential oils that are recently known for its applicability in food and pharmaceutical industry. The main problems of essential oil are most of the component likely unstable and also very volatile. Microencapsulation is a method whereby one material or a mixture of the materials is coated by another material. This method is designed for protection, isolation and assist in storage. In the present study, the gelatine-chitosan microcapsules were prepared by complex coacervation. In this study, encapsulated citronella oil was placed in simulated floor cleaner stimulated (Tween 80 solution).On the optical microscope, the microcapsules were spherical in shape and of irregular size. This microcapsule is confirmed by analysis of IR spectra of release of citronella oil in simulated clean floor solution. Release rate of the microcapsules (closed container) at different stirring rate at the beginning is slowly at 12 hours and begin burst and rapidly after 48 hours. When stirring increase from 300 rpm to 700 rpm the release rate increase to 80% in two weeks. Release rates of the microcapsules (closed container) at different temperature begin with a high rate in the first five days had become slow and stable after ten days. The microcapsules release rates were at 42%, 62% and 68% at 25°C, 50°C, and 75°C, respectively after 30 days. As conclusion the encapsulation of citronella oil by complex coacervation with gelatine and chitosan as core materials will improve the quality of the microcapsules. Release rates of the microcapsules at different conditions also indicate that the released rates were influenced by the stirring rate, temperature, and closed condition.

Keyword: Complex coacervation , control realease , simulated clean floor solution

1.0 Introduction

Citronella essential oil is acquired from the plant family of the Cymbopogon genus. More exactly, the grasses Cymbopogon nardus (Jowitt) and Cymbopogon winterianus (Rendal) are extracted by steam distillation. Citronella oil is also commonly used in fragrances and products for personal care (V. Pattusamy et al., 2013). Citronella oil is also used in soaps, household cleaners and detergents due to its antiseptic characteristics. Other research has indicated that citronella oil has powerful antifungal characteristics that can assist supress fungal species development, such as Aspergillus, Penicillium and Eurotium. To compound such as methyl isoeugenol, Citronella oil owes its antibacterial and antiseptic nature. These compounds stop bacteria from growing in the human body and help treat injuries and diseases that can happen in the colon, urethra,

bladder, gastrointestinal tract, prostate, and kidneys (P. Subramaniam et.al., 2015 and O. Avoseh et al., 2015). Since most essential oils are volatile, therefore, encapsulation should be introduced to control the process of evaporation (S. Varona et al., 2009).

Lately, researchers have been investigating the encapsulation of essential oil, since EO is easily evaporated. Encapsulation has the ability to block the biological activity of 33fragile essential oil by volatilization or degradation of effective components at elevated temperatures, UV and oxidation. Microencapsulation and nanoencapsulation are types of encapsulation used for essential oils. Preparation of encapsulation can be done by coacervation technique. Coacervation is the separation method of two liquid phases in colloid solution, whereby one phase is rich in polymer or coacervation phase, the other phase being free of polymer or equilibrium solution (A. EL Asbahani et al., 2015, F. Xing et al., 2005 and P.V Gadkari et al., 2013). To secure a long shelf-life for the essential oil, controlled release is the best way to achieve this. Controlled release can be measured by the amount of essential oil that can be released in the medium or environment after it has been encapsulated (C.Guo et.al.,2013). In this research, the encapsulation of citronella oil with gelatine-chitosan and glutaraldehyde as cross-linker was investigated. The morphology and release behaviour of the encapsulation of citronella oil (ECO) were first identified before proceeding to release the properties of ECO. A simulation is an approximate imitation of the operation of a process or system; that represents its operation over time.

2.0 Problem Statement

Essential oil such as citronella oil is known to possess antibacterial properties. The essential oil extracted from the leaves contains eugenol, a phenolic compound that may be attributed to its antibacterial properties. From the previous study, researcher had only focused on the reaction of citronella oil to a specific bacterium such as Enterococcus faecalis and not to other bacterial (P. Subramaniam.et.al., 2015). Recent clean floor in the market contains only few additive that has antiacterial agent and some of the additive contains chemicals that can harmful. Hence, application of citronella oil in cleaning solution must been carried out. CO is easily vaporized; thus, to overcome this problem, encapsulation process is introduced (P.T.Da Silva et al.,2014). Complex coacervation by using gelatine- chitosan might increase the encapsulation efficiency and loading capacity compare other coating material (Mumanya Mishra, 2016). The release mechanism of the microencapsulation citronella oil can identify before measure the release rate of the microcapsule (Z. Dong et al.,2011). Study shows that less research has been done to investigate the controlled release of citronella oil in different condition such as stirring rate and temperature after the encapsulation process and no work has been done on its application in floor cleaning.

3.0 Objective of study

Objectives of this study are firstly to encapsulate CO using complex coacervation technique, with gelatine and chitosan as a coating material. Secondly, to characterize the encapsulated CO on its morphology. Finally, to evaluate the controlled release behaviour of encapsulation of citronella oil (ECO) in a stimulated floor cleaning solution (Tween 80 solution).

4.0 Literature review

Encapsulation is described as a technology for enclosing solids, liquid or gaseous or miniature closed capsule products which vary from a nanometer to a micrometer spectrum which can discharge their contents under particular conditions at a regulated pace (Mumanya Mishra,2016). This method relies on the product to be encapsulated's physical and chemical properties. Microencapsulation technology has been used in a variety of industries such as chemicals, cosmetics, food, pharmaceutical or printing. The capsule has the capacity to maintain and discharge a product as necessary in a finely split state. The size of the capsules can vary from a submicrometer to several millimeters in size and have many distinct forms based on the components and techniques used to prepare them. The active ingredient encapsulation is performed for a multitude of purposes (G.Orive et.al.,2004).

The main reasons are protecting the core material from degradation by reducing its reactivity to outside environment such as UV light, heat, moisture, oxidation, etc. Other reason such as reducing or retarding the evaporation or transfer rate of volatile active ingredient to the outside environment. Also achieving controlled or targeted release of active ingredient whereby the product can be tailored to either release slowly over time or at a certain point also the reason for encapsulation (F.V Lemann et.al.,2009). Microcapsules can be categorized into three types such as encapsulation of mononuclear, polynuclear and matrix (Mumanya Mishra,2016). Also recognized as core cell microcapsules, mononuclear contains the shell around the heart as well as a single nucleus or monocore capsule.

Polynuclear capsules contain many cores within the shell, which is also called capsules of the polycore or multicore sort. Matrix encapsulation where the key material within the shell structure is divided homogeneously. In the senses, the microencapsulation method effectively includes on a time scale three distinct processes. The first method consists of forming a key material shell wall. The second process includes maintaining inside the wall fabric the key components intact so that they do not loose.

The third method includes releasing at the correct moment and speed the key content (Mumanya Mishra,2016). Microencapsulation method choice is determined by the heart and shell or fabric materials 'physical and chemical

characteristics and the desired implementation.

To design microcapsules with a broad range of functionalities, various techniques and shell material have been created (Mumanya Mishra,2016). By using selective encapsulation techniques and shell materials, designed microcapsule with controlled and or targeted release of the active encapsulated ingredients by using triggers such as pH change, mechanical stress, temperature, enzymatic activity, etc. can be obtained. Coacervation, cocrystallization, emulsion polymerization, spray drying and solvent evaporation are the example of microencapsulation process (A. EL Asbahani et.al.,2015).

The most important process is coacervation which are used widely in natural product. Coacervation is a small spherical droplet of assorted organic molecules, kept together by hydrophobic forces and varying in size from 1 to 100 µm (A. EL Asbahani et.al.,2015). There are two types of coacervation process that are easy and complex. Then solution of hydrophilic colloid are mixed under suitable condition. The microcapsules are usually collected by filtration or centrifugation and washed with the appropriate solvent dried by standard technique such as spray (A. EL Asbahani et.al.,2015.

To be encapsulated, the flavour or component is added to the carrier and homogenized to generate a tiny droplet. The resulting emulsion is supplied into the spray dryer's warm room where a nozzle atomizes it. Hot air contacts the atomizes droplet, the shell material solidifies onto core particles as the solvent evaporates and leaving dried particles (B.N.Estivinho et.al .,2005). After the encapsulation process the core ingredient were been released slowly and these released can be called as control release. Microcapsule morphology is the most important component of assessing whether or not the method of encapsulation is successful. The optical microscope and scanning electron microscope (SEM) are two popular methods used by the investigator.

The implementation and intent of both types of these microscopes were distinct. Controlled release is a term referring to the presentation or delivering of the compounds in response to stimuli such as pH, enzymes, light, magnetic fields, temperature, ultrasonic, osmosis and more recently electronic control or time. It may be defined as a method by which one or more active ingredients are made available at a desired site and time at a specific rate (M.A.Manaf et.al, 2014).

This terminology commonly refers to time dependent release, sustained release, pulse release, delayed release in oral dose formulations. There are many advantages as well challenges such as biocompatibility, the fate of controlled release system if not biodegradable, cost of formulation, etc. associated with the designing a controlled release system or formulation. Other encapsulation technologies including enteric coating can further modify release profiles. Microencapsulation is a technology than control dissolution profiles. The dissolution rates can be further controlled by further coating and layering the microcapsule or microsphere with insoluble substances (W.C.Hsieh et.al .,2006).

5.0 Materials and methods

5.1 Material preparation

The materials used as wall materials in this research were gelatin-B (type B, 260 bloom, from bovine) from Halagel Sdn. Bhd and chitosan medium molecular weight from R&M Chemical. The essential oil used as a core material was citronella essential oil (CO) of lemongrass type java 85/35 % from Sigma Aldrich, which was supplied from Aman Semesta Sdn. Bhd. Glutaraldehyde (50% aqueous solution) from Merk was used as a cross-linker. Other reagents involved are distilled water, ethanol, acetic acid solution (1ml acetic acid in 100 ml of distilled water) from R & M, hydrochloric acid (HCl) 0.5 -50% from R & M, and sodium hydroxide (NaOH) 99 % from R & M.

5.2 Encapsulation Procedure

Before entering the encapsulation process, two main solutions were prepared; solution A and solution B must first be done. Solution A was prepared by dissolving 3.5 g of gelatine in 350 g of deionised water. The solution A was stirred for one hour at 50°C until the solution was fully dissolved. Solution B was prepared by dissolving 0.1 g of chitosan in 100 g of acetic acid aqueous solution (0.1 wt %) for more than 12 hours at room temperature. The method was adopted by F.R.Abdul Aziz .et.al.,2016.

There are seven steps involved in complex coacervation. The microencapsulation process is shown in **Figure 1**, where the first step is the emulsification. In this step, 3.6 g of CO is added into solution A. This CO will be the core material for coacervation. The solution is stirred at 500 rpm for 30 minutes. The second step is the coacervation. In this step, a 100 ml of solution B is added drop by drop into solution A. The mixture is maintained at 50°C for 30 minutes with a pH of 5 and stirred at 500 rpm. This kind of process for emulsifying oil in an aqueous solution containing two different polymers.

The third step is the pH adjusting. The mixture is adjusted to a pH of 5 by adding 0.1M of NaOH or CH3COOH, drop by drop. The mixture is maintained at 50°C and stirred at 500 rpm for 30 minutes. The fourth step is the dilution. A 200 ml of distilled water is added into the mixture solution and the mixture is maintained at 50°C and stirred at 500 rpm for 30 minutes.. The fifth step was the cooling process; a 200 ml of distilled water is added into the mixture solution. The mixed solution is kept in the ice bath, maintained at 10°C and stirred at 200 rpm for 60 minutes (F.R.Abdul Aziz .et.al.,2016).

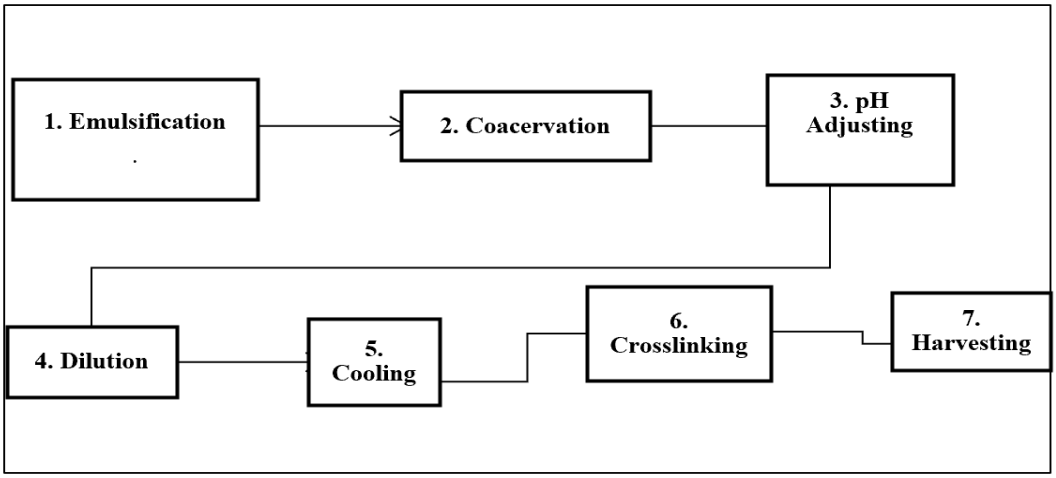


Figure 1: Microencapsulation process using Gelatin- Chitosan

5.3 Control Release Behaviour of ECO

Controlled release of the ECO was determined by identifying the percentage release of ECO under specific time taken. The controlled release study was done using the UV-Vis spectrometer. From the UV-Vis spectrum, the release profile of ECO can be determined. The factors affecting the percentage release of ECO, such as the stirring rate and temperature, were taken into account. The objective of this study was to find the standard calibration curve for citronella oil in the determination of unknown sample from the citronella oil solution. A known concentration of citronella in 100 ml of distilled water containing 0.3% Tween 80 was scanned in the range 200–400 nm using UV-Vis spectrometer, Perkin Elmer, Lambda 750, as shown in Plate 1. By using 0.3% Tween 80 as blank then, one gram of CO was dissolved in the simulated floor cleaner Tween 80 of 0.3% w/v. For CO having a blank concentration from 5–100 µl, a sharp peak at 333 nm was noticed. The absorbance value at 333 nm was obtained, with its respective concentration recorded and plotted. From the calibration curve, the unknown concentration of CO was obtained by knowing the absorbance value (Maji et.al.,2007). The total amount of CO in the microcapsules was identified by grinding it with blender (Pensonic PB-3203l) to fine particles and crushing a certain weight of ECO with mortar and pestle prior to having it dissolved in the simulated floor cleaner while keeping it stirred for 24 hours at 50 rpm. The solution was tested by using the UV spectrometer to determine the concentration of the released CO through comparison of the standard calibration curve.

Plate 1 : UV-Vis Spectrometer, Perkin Elmer, Lamda 750

The release of CO was determined by measuring the solution of the stimulated floor cleaner with different concentrations of ECO, where the measurement was done using UV-Vis (Maji et.al.,2007) . In order to determine the concentration of the sample by comparing the standard calibration curve, the amount of citronella oil on the percentage release of the microcapsules was calculated according to the formula below:

Release percent (%) = ($\frac{C_0}{C_d}$) x 100 % Where Cd is the total amount citronella oil in the microcapsules and C0 is the amount of citronella oil in the microcapsules at the test date. This method was adopted by Maji.et.al.,2007

5.4 The release percent ECO at closed container at different temperature.

The main purpose of this study is to find the percentage release of ECO in a solution in closed container. The closed container at different temperatures represents the condition where the floor cleaner is kept before being used by consumers. The release behaviour of ECO was determined by conducting a series of experiment with different weight (0.5, 1.0 and 1.5 g) of ECO in simulated floor cleaner at different temperatures (25°C, 50°C, and 75°C). The sampling was done periodically at an interval of 5 days. **Table1** shows the sample with different concentrations of ECO due to the varied temperature in the close container (Maji et.al.,2007).

Table 1: The different weight of ECO in 0.3 % Tween 80 at varies temperature for closed container.

Weight of ECO in 0.3 % w/v Tween 80	Varies of Temperature for Closed container		
	25⁰ C	**50⁰ C**	**75⁰ C**
0.5 g	Sample 1	Sample 1	Sample1
1.0 g	Sample 2	Sample 2	Sample 2
1.5 g	Sample 3	Sample 3	Sample 3

5.5 Effect of Stirring on Release Rate of ECO (closed container)

The main purpose of this study is to investigate the effect of different stirring rates on the release rate of ECO in the solution. The closed container with stirring effect represents the condition when the stored floor cleaner undergoes vigorous state. The release behaviour of ECO were determined by conducting a series experiment with different concentrations of ECO in simulated floor cleaner at different stirring rates (300.500 and 700 rpm) in the interval of two weeks. **Table 2** shows the sample with different concentrations of ECO due to the varying stirring rates in closed container (Maji et.al.,2007).

Table 2: The different weight of ECO in 0.3 % Tween 80 at varies stirring rate for closed container.

Weight of ECO in 0.3 % w/v Tween 80	Varies of Stirring Rate for Closed container		
	300 rpm	**500 rpm**	**700 rpm**
0.5 g	Sample 1	Sample 1	Sample1
1.0 g	Sample 2	Sample 2	Sampel 2
1.5 g	Sample 3	Sample 3	Sample 3

5.6 Release rate of microcapsules in open container

The main purpose of this study is to investigate the release rate of ECO in the solution at open airs. The open container represents

application of floor cleaner on the floor. About 0.5 g of the weight of microcapsules was placed in a four-petri dish containing 20 ml of a solution containing different concentration of ECO in simulated floor cleaner. The petri dish was open to room temperature. Every each day the microcapsules were taken from the petri dish. The release behavior from CO were determined by conducting a series experiment with different concentration of ECO in simulated floor cleaner at in two weeks .This method was adapted from Maji.et.al. Table 3 shows the sample with the different concentration of ECO in stimulated floor cleaner at open container .

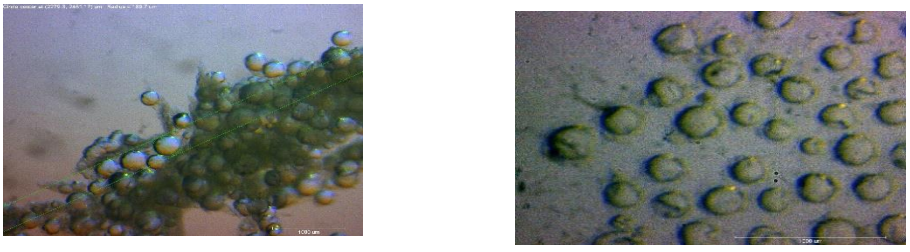
Table 3: The different weight of ECO in simulated clean floor at open container

Different weight of ECO in 0.3 % w/v Tween 80	Type of sample
0.5 g	Sample 1
1.0 g	Sample 2
1.5 g	Sample 3

6.0 Discussion

6.1 Shape and morphology of ECO

The encapsulation of citronella oil by complex coacervation and gelatin-chitosan as core shield was successful. **Figure 2** shows the microcapsules of citronella oils under an optical microscope.



(A) Microcapsules in solid forms agglomerates (B) Microcapsules dispersed in distilled water

Figure 2*:* The microscopic image of the microcapsules under the optical microscope

Figure 2 shows that the CO capsules are in a spherical shape, which has

irregular size. Some of the capsules were attached together, causing agglomeration due to the hardened free gelatine–B after cooling until the settling process. The agglomeration of the capsules during the wall formation was a common phenomenon in many microencapsulation processes(F.R.Abdul Aziz .et.al.,2016). As the wall of the materials changes from liquid to solid form, they often undergo a sticky stage, which makes the agglomeration difficult to avoid (Nack.H ,1970) (Mumanya Mishra,2016). After the microcapsules were dispersed in the water, the spherical microcapsules can clearly be seen, as shown in **Figure 3** below.

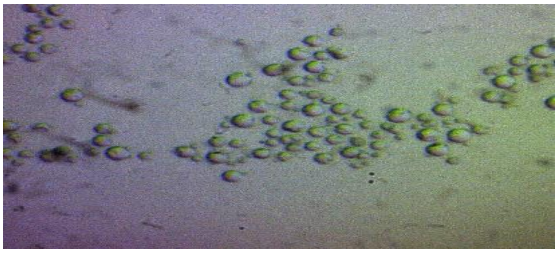


Figure 3 :The picture of the microcapsules dispersed in clean floor solution.

The release of microcapsules can be observed by dispersing ECO in the clean floor solution for two weeks. **Figure 4** shows the release of ECO in clean floor solution This figure shows that the wall of the microcapsules had slowly ruptured on day six in the stimulated clean floor and after two weeks, it completely ruptured and the oil was fully released from the microcapsules.

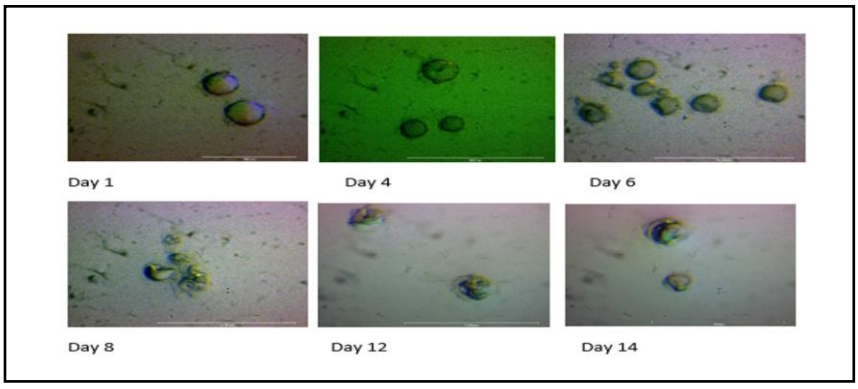


Figure 4 : The picture of the microcapsules of CO rupture in 0.3 % Tween 80 solution.

6.2 Effect of release percent of CO (closed container) at different stirring rate.

The release of citronella oil through the microcapsules in 0.3% Tween 80 of aqueous solution could be divided into two phases: citronella oil released before 12 hours and after 48 hours, where the burst and rapid release of the citronella oil could be observed. **Figure 5** shows the release rates of microcapsules at different stirring rates. At the stirring rate of 300 rpm, the profile release of microcapsules shows that the percentage release was smaller than microcapsules stirred at 500 rpm and 700 rpm. It could be identified by looking at the final release of the microcapsules. The final release for microcapsules had increased more than 80% when the stirring rate was increased from 300 rpm to 700 rpm. It might have been because when the microcapsules were slowly stirred, the microcapsules' core shield, such as gelatine and chitosan, were fully protecting the microcapsules from being easily ruptured and releasing the oils. When the stirring rate was increased to 700 rpm, a strong force had been applied to the microcapsules and it had weakened the core shield of the microcapsules to rupture even more and increase the release of oil into the solution. Similar finding has been observed by other researcher (Z.Dong et.al., 2011)(L.F. Siew et.al.,2012). The different amount of microcapsules did not give much effect on the release rate of the microcapsules. This can also be seen in *Figure 4.3*, whereby the release pattern for microcapsules of 0.5 g, 1.0 g and 1.5 g at three different stirring rates were almost the same. This is because the concentration of the microcapsules is insignificant to the release rate of the microcapsules. The factors affecting the percentage release are related to the interaction between the wall and the core material. From this analysis, it also shows that when the weight of microcapsules and stirring rate were increased, the release rate had also increased. The result reveals that the release rate of CO for all stirring rate under study was not effected by the concentration of ECO. The graph pattern of release rate was classified as following the Korsmeyer-Peppas controlled release model of super case II, whereby the release is due to wall erosion.

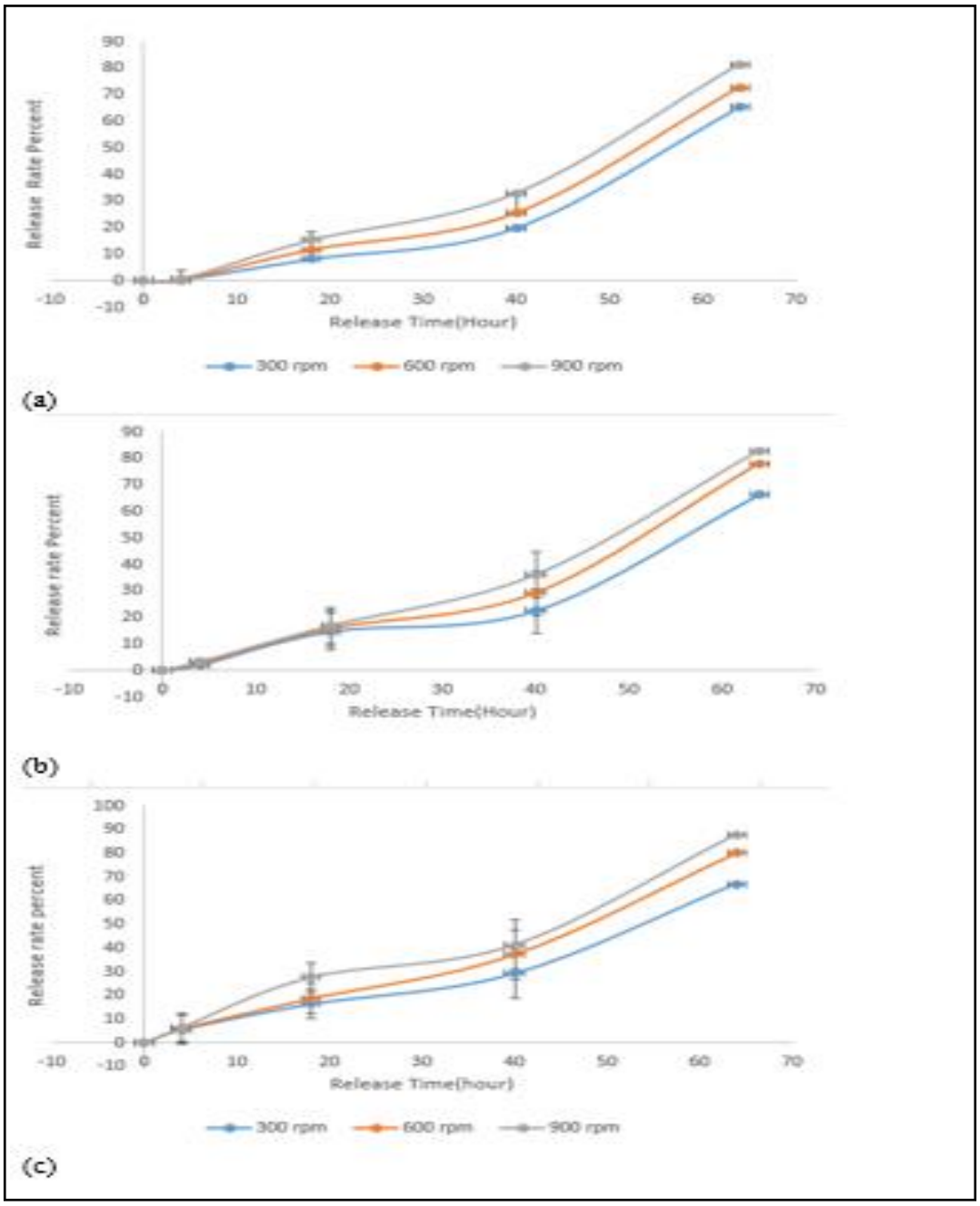


Figure 5 : The effect of release percent of ECO on diferent stirring in closed container

6.3 Effect of release percent of microcapsule at different room temperatures (closed container)

The release profile of different microcapsules containing citronella oil in 0.3% Tween 80 aqueous was studied at 25, 50 and 75°C. As shown in **Figure 6** the citronella oil released from the microcapsules at a high rate within the first five days had become slow and stable after ten days. The reason might

be because the citronella oil was distributed on the surface or surface layer, which was easier to release in the primary release stage; thus, the cumulative release rate was higher within the first release stage. This is applied to all of the different weights of the microcapsules used in this experiment. Once the citronella oil on the surface layer is released completely, the next stage would remain relatively stable due to citronella oil in the microcapsules that were mainly released through the microcapsules' wall through the penetration effect (P.T.Silva et.al.,2014).

After 30 days, the citronella oil still existed in the microcapsules and the microcapsules release rates were at 42%, 62% and 68% at 25°C, 50°C, and 75°C, respectively. That is, the citronella oil in the microcapsules could still be remained at 58% at 25°C after its release for 30 days. The main reason might be due to the vapour pressure of citronella oil that had increased, leading to the increased volatility of the citronella oil. As confirmed by the experiment, the stability and storage period of the citronella oil were greatly enhanced, while achieving durable controlled release effect and long residual action through microencapsulation (Z.Yang et.al.,2014)]. Similar finding by the other researcher claimed that the percentage release of the microcapsules had increased when the temperature was increased and the duration of time release shorter than the release rate of the microcapsules at normal condition (W.C.Hsieh et.al.,2006). Microcapsule weights of 1.0 g and 1.5 g showed that the percentage releases were higher compared to the 0.5 g of microcapsules when the temperature was increased. The result also reveals that the release rate of CO for all temperatures under study was not affected by the concentration of ECO. The graph pattern of release rate was classified as following the Korsmeyer-Peppas controlled release model of super case II, whereby the release is due to wall erosion.

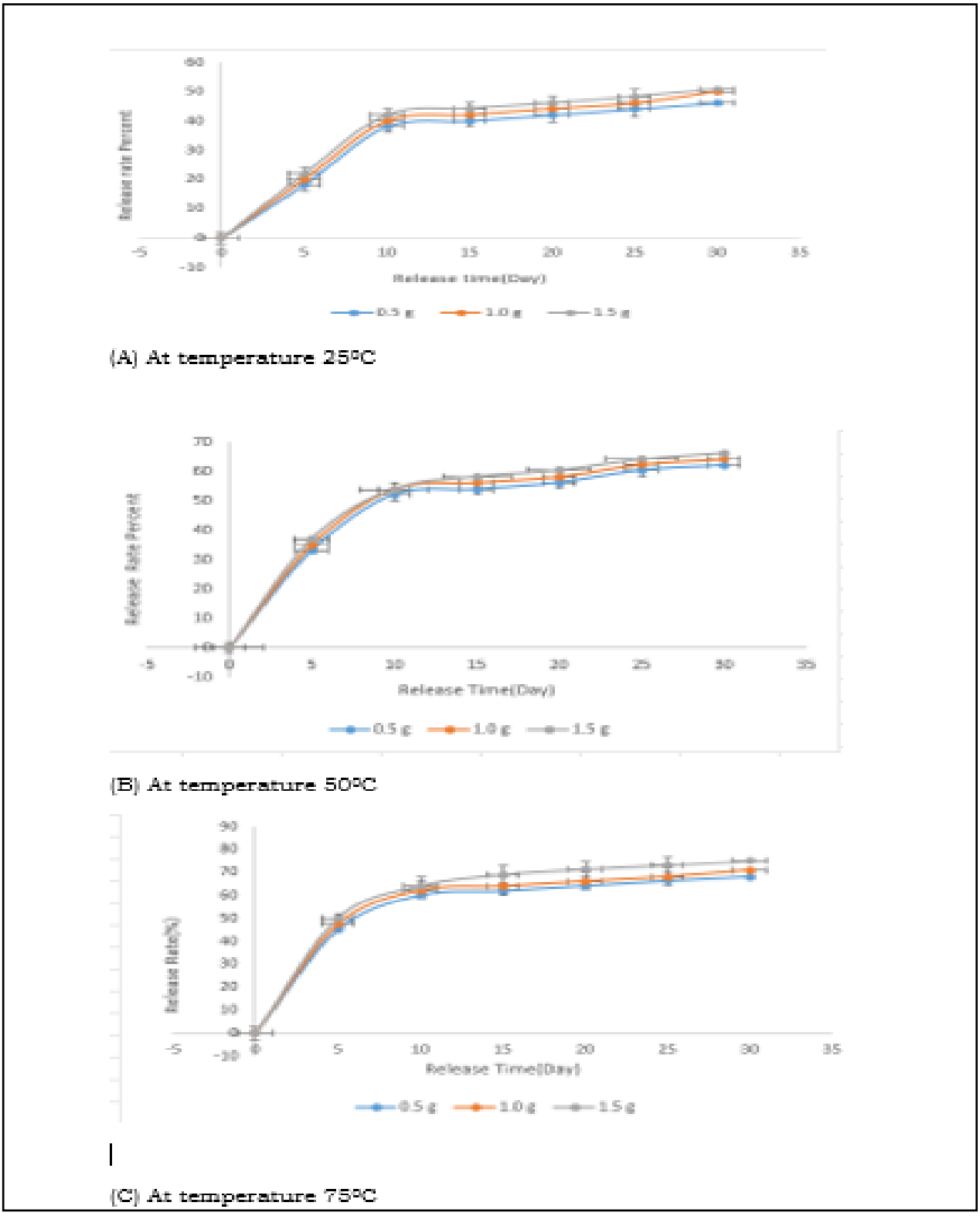


Figure 6: The effect of release rate microcapsules at different temperature (closed container)

7.0 Conclusion

The encapsulation of citronella oil by complex coacervation with gelatine and chitosan as core materials were successful. The microcapsules produced with the CO capsules were in spherical shape, having an irregular size. Controlled release of ECO shows at normal condition, the release rate is slow and the release rate was able to withstand up to three months until it achieved a full release of 100%. The effect of stirring rate and temperature had altered the release rate of CO in solutions such as 0.3% Tween 80 and methanol. The release rate of microcapsules had increased when the stirring rate was increased from 300 rpm to 900 rpm. Within one week the ECO were fully released .The release rate of microcapsules had also increased when the temperature of the solution containing ECO increased from 25°C to 100°C. The CO was released from the microcapsules at a high rate within the first five days, and it became slow and had a stable release after ten days. The duration of full release of microcapsules was within two months. In the future the release rate of microcapsules can be identified by using a fluorescence spectroscopy. Fluorescence is a rapid, sensitive method for characterizing molecular environments and events. There are a relatively small number of compounds that have a characteristic fluorescence, such as aromatic hydrocarbons. This has contributed to the successful use of the spectroscope in the detection of many of organic compounds, aromatic numerous active substances in drugs in the field of research chemicals

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