UNIVERSITI PUTRA MALAYSIA

SHELF LIFE COMPARISON STUDY OF ENCAPSULATED AND NON-ENCAPSULATED JASMINE FLOWER'S EXTRACT

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ABSTRACT

Jasmine fragrance consists of thermal-sensitive chemical compounds such as linalool, farnesene and benzyl acetate that are volatile and easily degrade. Therefore, non-thermal solidification method - electrospray is preferred when compare to the thermal-assisted atomization technique i.e spray drying that can cause thermal degradation of the atomized flower's compounds. Electrospray is a method of liquid atomization by means of electrical force that involved the mechanisms of solvent evaporation and coulomb fission in order to produce solidified jasmine's compounds powders. The parameters of jasmine compound concentration (wt%) in water and relative humidity of surrounding are varied in order to determine the drying rate of the atomized droplet . The studied indicated that the drying rate is proportional to the psychrometric properties of the electrospray system. Prior to the sensitivity of the chemical compounds, the atomized droplets are encapsulated with β-cyclodextrin (β-CD) and the effectiveness of the electrospraying process to solidify the droplet is compared with the non-encapsulated ones. The field Emission Scanning Electron Microscopy (FE-SEM), Fourier Transform Infrared Spectroscopy (FTIR) and thermogravimetric analysis (TGA) were used to verify the occurrence of encapsulation. Morphology of the encapsulated bioactive compounds was found to be in the spherical crystal lattice from and free from agglomeration. The FTIR also showed that the encapsulated droplets were consisting of identical peaks of β-CD and extracted jasmine compounds that indicates the occurrence of the encapsulation process. In the TGA curve, the shifting of thermal degradation of the encapsulated droplet to higher temperature profile compared to the non-encapsulated ones also verified the occurrence of inclusion complex between the compounds and the β-CD. The shelf life and stability

of encapsulated jasmine compound and non-encapsulated ones was compared in the close system storage for 28 days. The loading capacity of the encapsulated bioactive compounds was found to be decreasing week by week but the difference is not very significant. As a conclusion, this study is important to be carried out in order to find an effective method to enhance and prolong the shelf life of the bioactive compounds using the electrospray process.

ABSTRAK

Wangian bunga melur (Jasmine) terdiri daripada sebatian kimia sensitif termal seperti linalool, farnesene dan benzyl acetate yang tidak menentu dan mudah dibebaskan apabila terdedah kepada tekanan haba. Oleh itu, kaedah peneringan bukan haba iaitu pengeringan semburan electro dipilih apabila dibandingkan dengan teknik pengeringan semburan dengan menggunakan haba. Disebabkan sensitiviti sebatian kimia, titisan atomized dikemas dengan β-siklodekstrin (β-CD) dan keberkesanan proses semburan electro untuk menguatkan titisan berbanding dengan yang tidak terkandung. Pengeringan semburan electro adalah kaedah pengabusan cecair dengan menggunakan kuasa elektrik yang melibatkan mekanisme penyejatan pelarut dan '*Coulomb fission*' untuk menghasilkan serbuk bunga melati. Parameter kepekatan (wt%) sebatian melati dalam air dan kelembapan di persekitaran yang berbeza untuk mendapatkan kadar pengeringan. Ia menunjukkan bahawa kadar pengeringan meningkat dengan kelembapan. Kemudian, Mikroskopi Pengimbasan Pelepasan Medan (FE-SEM), '*Fourier Transform Infrared Spectroscopy*' (FTIR) dan analisis termogravimetrik (TGA) digunakan untuk mengesahkan kejadian enkapsulasi. Morfologi sebatian bioaktif yang terkandung didapati dalam sfera dan tidak aglomerat manakala dalam FTIR, sebatian bioaktif yang terkandung mempunyai puncak yang sama dari β-siklodekstrin dan bunga melati yang diekstrak menunjukkan enkapsulasi berlaku. Dalam TGA, kemerosotan sebatian bioaktif kepada suhu yang lebih tinggi dalam enkaspsulasi menunjukkan bahawa kompleks inklusi telah berlaku. Kestabilan sebatian melati yang dienkapsulasi telah dibandingkan sebelum dan selepas penyimpanan 28 hari. Kapasiti memuatkan sebatian bioaktif kian menurun tetapi perbezaannya tidak

begitu ketara. Sebagai kesimpulan, kajian ini penting untuk dijalankan untuk mencari kaedah yang berkesan untuk meningkatkan dan memanjangkan jangka hayat sebatian bioaktif.

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LIST OF SYMBOLS

EHDA	Electrohydrodynamic atomization
CD	β-cyclodextrin
FESEM	Field Emission Scanning Electron Microscopy
FTIR	Fourier Transform Infrared Spectroscopy
TGA	Thermogravimetric Analysis
DTGA	Derivative Thermogravimetric Analysis
$\frac{dw}{dt}$	Mass being transferred (kg/s)
k	Mass transfer coefficient (kg/m^2s)
A	Area through which the transfer is taking place (m^2)
ΔY	Humidity difference (kg/kg)
RH	Relative humidity
d_0^2	Initial droplet diameter (m^2)
d_p^2	Diameter of particle deposited (m^2)
t	time (s)
k	Evaporation rate constant
IEC	Initial encapsulated compound
FEC	Encapsulated compound after 28 days of storage
EBC	Encapsulated bioactive compound
NBC	Non-encapsulated bioactive compound

CHAPTER 1

INTRODUCTION

1.1 Background

Jasmine with scientific name *Jaminum* in the Family Oleaceae which is a small tree mostly cultivated in warm temperature region and bear with tiny white flower that possess unique and enthusiastic aromas. According to Pragadheesh et.al (2017), jasmine flower possesses a powerful fragrance and the essential oil was possessed antibacterial activity. Flower aroma is widely different in terms of their identity and relative amount of volatile constituent for example, volatiles constituents that present in Jasmine flower extract including linalool, benzyl-acetate and farnesene. Among these, linalool is the major flora scent in nature (Knudsen, 2006) but in this study, the constituents of jasmine flower extract were expressed as bioactive compound instead of focus on particular components. Therefore, incorporation of these beneficial bioactive compounds into food or drug in order to improve its quality have raised the interest among food and pharmaceutical industry. However, these compounds have high volatility and limited water solubility thus it was challenging to retain the presence of bioactive compounds in consumers' product (Elia et al., 2015). As a result of these drawbacks, the protection and controlled release of bioactive compounds can be implemented by encapsulation.

Encapsulation offer many advantages such as to cover an undesirable flavour, smell or taste, to control the release, to change the physical properties of the initial substances and to surpass the bioavailability of bioactive compound (Fakhreddin, Zandi, Rezaei, & Farahmandghavi, 2013). Encapsulation is a technique by which one material or a mixture of materials is coated with or entrapped within another material or system. Encapsulation of flavour and fragrance have been commercialized to prevent retardation by using many techniques including spray drying, spray cooling, coacervation, fluidized bed coating, extrusion and inclusion complexation (Risch, 1995). Spray drying is the most established well-known technique for drying during encapsulation because of low cost, flexible and lead to production of stable and high quality product (Munin & Edwards-Lévy, 2011). However, this technique is required high temperature operating condition that is not suitable for temperature sensitive compound such as phenolic compounds. Coacervation is a well-known technique for encapsulation as well, but it is considered as an expensive technique for encapsulation of food ingredients and usually associated with no definite forms (Gouin, 2004). Due to the drawbacks of these techniques, an alternative technique has been explored which is electrohydrodynamic atomization (EHDA). This technique has been employed in a variety areas, for example, fuel injection, ink-jet printing and manufacture microparticles for drug delivery.

Table 1 Comparison between thermal and non-thermal drying methods (Bhushani, Kurrey, & Anandharamakrishnan, 2017)

Operating Parameters	Thermal	Non-thermal

Example	Spray drying	Electrospray
Temperature	Involved high inlet and outlet temperature	Non-thermal process; ambient room temperature
Morphology of particles	Spherical, dimpled, hollow	Spherical with fine pores
Particle size	Heterogeneous particle size due to aggregation; microparticles	Mono dispersed, non-aggregated; nanoparticles
Product quality	Denaturation may occur for heat sensitive bioactive compounds	No denatured of proteins or bioactive compounds
Encapsulation efficiency	Medium to high	High
Energy consumption	High energy required	Low energy required

In order to prolong the extracted bioactive compound, it has to be encapsulated with a coating agent to prevent compounds released or loss to surrounding in a short time (Madene, Jacquot, Scher, & Desobry, 2006). In this study, β - cyclodextrin (CD) is chosen due to its special structural shape and some other characteristics such as CD has the ability to modify the guest molecule's characteristics. For example, CD managed to enhance the solubility of lipophilic 'guests' in water, control volatility and masking potentially adverse flavour (Pinho, Grootveld, Soares, & Henriques, 2014).

CD molecules have a truncated cone shape, with a hydrophobic interior cavity and a hydrophilic external surface that can increase its water solubility and suitable for formation inclusion complexation through non-covalent interaction (Lian & Huang, 2012). For the encapsulation of flavour compound, the wall material or the encapsulating agent must have no reactivity to the core materials. Other than that, the encapsulating agent must have the ability to give maximum protection to the active compound against the external factors. There are several coating agents used in different encapsulation techniques, for example, maltodextrin in coacervation, α and β cyclodextrin in inclusion complexation (Ezhilarasi, Karthik, & Chhanwal, 2013) which are widely used in the food industry to retain and protect volatile compounds. β - cyclodextrin was commonly used in pharmaceutical industry, agricultural, chemical and foodstuff due to its appropriate size cavity and easy accessible (Lian & Huang, 2012).

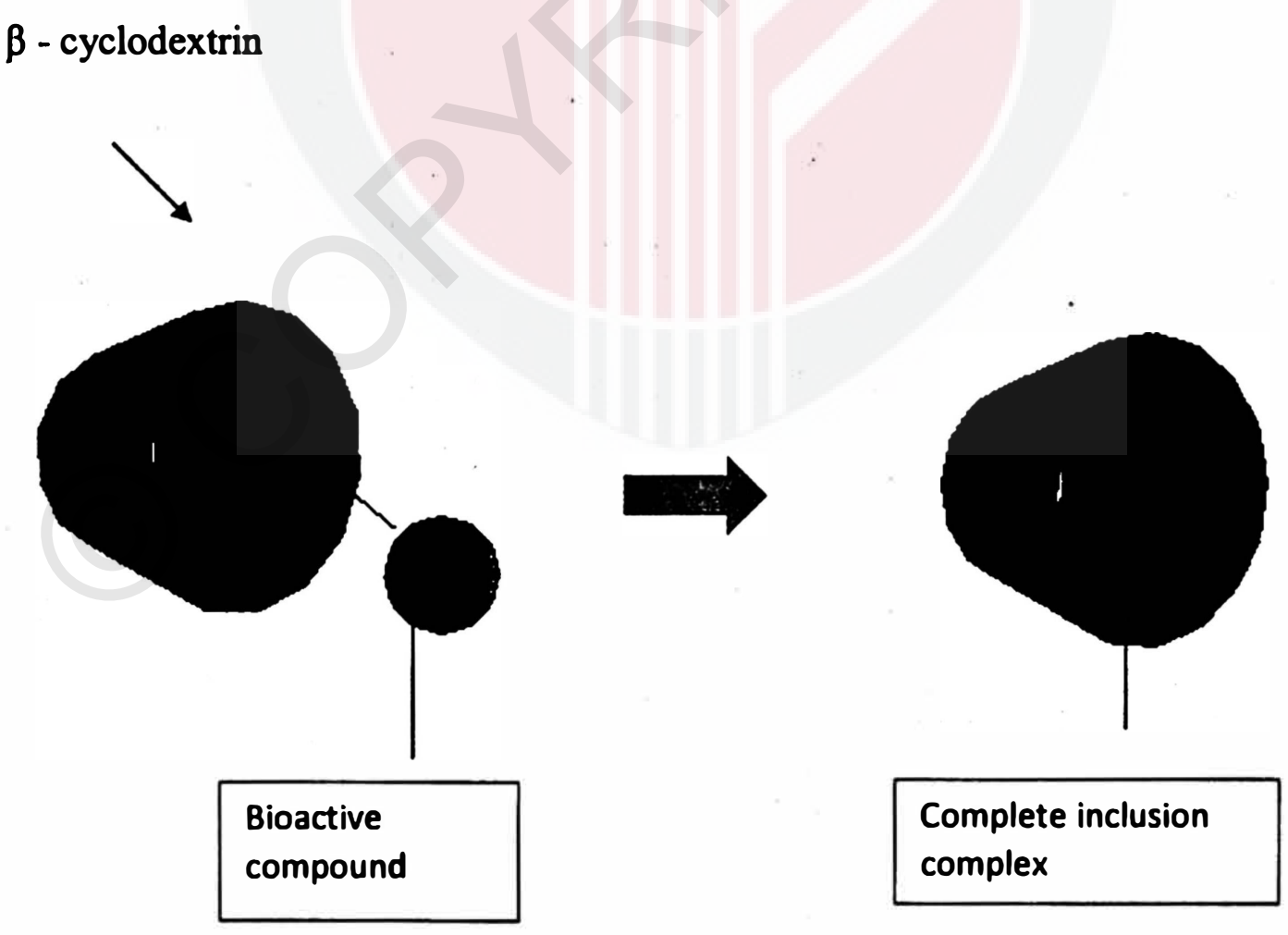


Figure 1 Schematic illustration of inclusion complex encapsulation of CD

Figure 1 illustrated the mechanism of the formation inclusion complex in which the proactive compound was encapsulated using β-cyclodextrin as a coating material. After CD complexation occurred, liquid substances can be converted into solid complex, thereby can improve its processing characteristics (Khan & Durakshan, 2013). The encapsulated compound contained solvent that need to be dry in order to get solidified particles. Commonly drying of encapsulated bioactive compound with CD required heat. One of the most common technology used in the food industry to produce encapsulated additives for food application is spray drying (Pérez-Masiá et al., 2015). The heating process in droplet solidification to produce nanoparticles tend to cause the encapsulated compound to degrade (Mphahlele, Fawole, Makunga, & Opara, 2016). Therefore, non-thermal drying by using electrospray techniques was chosen to encapsulate the bioactive compound and prevent them from quality changes.

Electrospray process is an electrohydrodynamic atomization in which the liquid feed is sprayed by the application of high potential electric field to obtain the particles which range from micrometer to nanometer particles. The electric potential difference applied between the capillary and a ground collector caused the droplets spray out from the capillary nozzle.

In order to generate nanoparticles in solid forms, several techniques can be applied either from solid, gas or liquid phase. In the case of solid phase, the material is crushed into fine particles whereas in liquid phase, nanoparticles can be generated from the atomization of liquid solutions and followed by the evaporation of solvent (Jaworek &

Sobczyk, 2008). In the gas phase, nanoparticles are produced from the condensation of vapours by chemical or physical means from a dissolve material (Rajput, 2015). In this case, atomization of liquid solution will be the main focus of the experiment.

During atomization, electrospray was started with getting a constant flow rate of droplets from the capillary nozzle. When an electric field was applied, it induces a surface charge in the droplets. As a result of electric stress, the droplet is transformed from dripping into a conical shape which is known as Taylor cone(Arya, Chakraborty, Dube, & Katti, 2009). A Taylor cone was developed due to surface tension alter by the high voltage static charge where the meniscus change from dripping mode into a jet mode. This jet will break into a number of primary or main droplets and numerous secondary droplets and satellites. Each surface of droplets was surrounded by static charge, when the static charge exceeding the Rayleigh limit, the droplet was unstable, thus caused the emission of water from droplet which is evaporation of water from the droplets (Bodnar & Rosell-Llompart, 2013).

During evaporation of water, the removal of water concentrated the charge per droplets causes the fission of droplet occur. The process of fission and water removal are continuity that will lead to a solidified or wet droplet. The solid particle still containing a minimum trapped charge that enables it to deposit on the collector which is applied with opposite charge. At the end of the electrostatic atomization process, different particle size distribution and surface morphology were collected at the aluminium. There are several parameters that affect the size and surface morphology of the droplets including the flow rate of liquid, the voltage applied at the nozzle, the distance

between the nozzle and the collector, temperature and humidity. This method has many advantages as it is a single step with low energy consumption and low cost material when compare to conventional spraying system such as spray drying. The charged spray has higher efficiency of the deposition of an object when compare with the un-charged spray (Jaworek & Sobczyk, 2008).

Finally, the atomized solidified particles were collected and compared between encapsulated and non-encapsulated with varies operation parameters. A drying rate curve based on the mass transfer equation was generated to determine the drying process using electrospray technique. Other than that, comparison of shelf life and stability of encapsulated and non-encapsulated JE at different concentration and distances were conducted and the suspension was solidified using electrostatic atomization technique. The solidified particles were analyzed and characterized by using Field Emission Scanning Electron Microscope (FESEM), Fourier Transform Infrared Spectroscopy (FTIR) and Thermal Gravimetric Analysis (TGA). The samples prepared via electrospray were analyzed and pattern of degradation of components of bioactive compound in Jasmine flower was compared before and after storage.

1.2 Problem Statement

Nano-particles can be generated when liquid suspension are disintegrated into fine droplets through atomization. Generally atomization can be divided into thermal and non-thermal atomization. The spray drying is using conventional atomization which involves thermal energy to evaporate the solvent in droplets that can destroy the bioactive compounds in Jasmine flower.

On the other hand, electrospray is one of the non-thermal atomization drying process and this technique has several advantages such as high encapsulation efficiency, greater thermal and storage stability and give spherical nanoparticle size (Bhushani, 2014). Electrospray can be carried out in atmospheric conditions which reduce the possibility of degradation and extend the shelf life of samples. Between, the mechanism of solvent evaporation and coulomb fission are used in the atomization process.

Other than that, the operation of electrospraying is cheap and the rate of particle produced can be controlled easily by varying the liquid flow rate and applied voltage (Jaworek & Sobczyk, 2008). However the parameters during electrospray drying might cause the incomplete encapsulation or crack formation that will affect the stability and shelf life of bioactive compound. Thus, how to effective encapsulation of bioactive compound into β-cyclodextrin still a very challenging and important issue in producing nanoparticles.

1.3 Objectives

1. To determine the drying rate of the atomized jasmine's extracts.

2. To identify the encapsulated compounds' shelf life with different drying rates after the atomization process.

1.4 Hypothesis

The beta-cyclodextrin which act as encapsulating agent will successfully formed an inclusion complex with bioactive compound in Jasmine flower to prevent degradation. Few studies have been done by several researchers on determination of inclusion complex between the aroma for example linalool and the beta-cyclodextrin. The complexation formation depends on the guest compound hydrophobicity as well as molecular size and geometry (Ciobanu et al., 2012)..

Electrospray atomization is used to produce nano size of particles without using thermal energy. Solvent evaporation and coulomb fission are the fundamental mechanism in electrospray drying process. Evaporation increases the surface charge density until it reached the Rayleigh limit leading to fission occur (Konermann, 2009). The condition during electrospray such as high humidity will cause the lower water evaporation of encapsulated and non-encapsulated Jasmine compound.

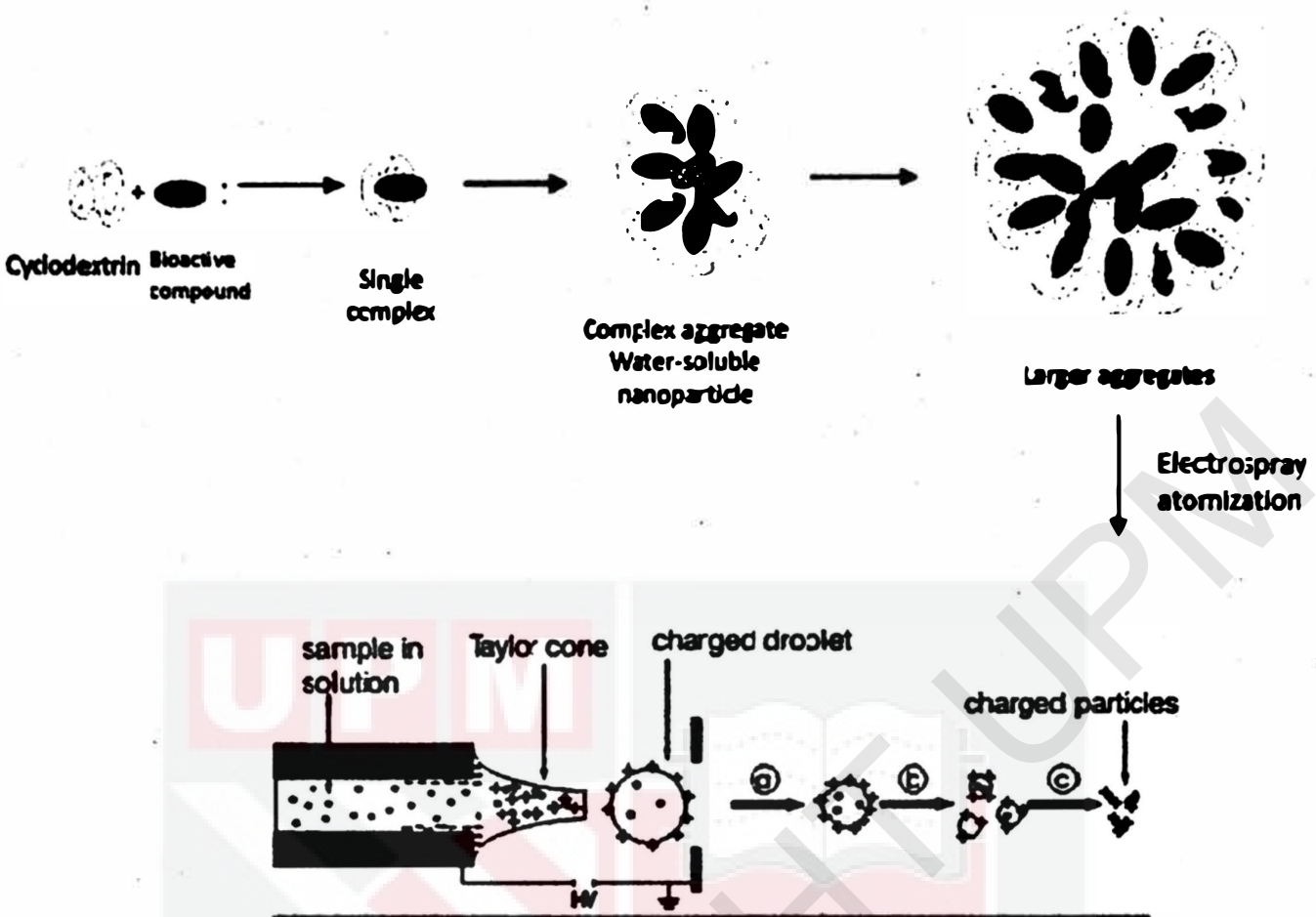


Figure 2 Illustration of complete encapsulation of CD with bioactive compound and electrospray atomization(Ryzhakov et al., 2016)

CHAPTER 2

LITERATURE REVIEW

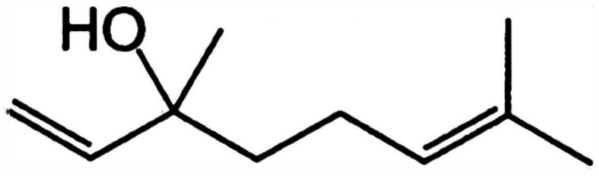
2.1 Bioactive compound

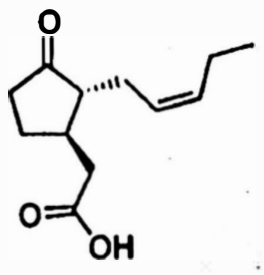
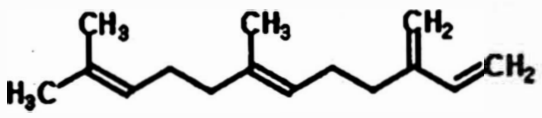
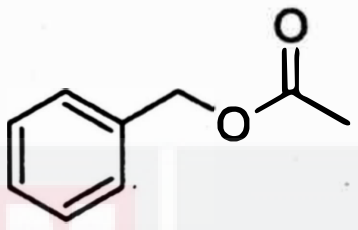
Bioactive compound (BC) is a compound which has the biological activity if it has a direct effect on a living tissue. These effects can be adverse or beneficial depending on the dose, substance or bioavailability. Bioactive compounds can be found in both plant and animal or synthetically produced. Example of bioactive compound that can be found in plants are carotenoids and polyphenols from fruits and vegetables. These components of bioactive compounds will face the stability problem either physical, chemical or biological when they were exposed to surroundings, for example Ferulic acid in whole grain wheat has a low stability to thermal and physical stress (Pinho et al., 2014) while tea catechins has low solubility in lipophilic substance which makes it difficult to be incorporated in lipid-based foods (Bhushani et al., 2017).

2.2 Bioactive Compounds in Jasmine

Jasmine (*Jasminum*) with local name Melati; Melur is known as white jasmine belong to Oleaceae family. Generally, floral scents consist of terpenoids, phenylpropanoids or benzenoids and fatty acid derivative compounds (Pragadheesh, Chanotiya, Rastogi, & Shasany, 2017). It is believed that Jasmine possesses an attractive fragrant among the flowers and their flowers has utilized as traditional treatment in Asia to cure many diseases such as skin disease, diarrhea, asthma, breast cancer and etc. Jasmine flower extract has a sweet and floral aroma which possesses powerful characteristics that aid the body in antidepressant while the antimicrobial activity (Kunhachan, Banchonglikitkul, Kajsongkram, Khayungarnnawee, & Leelamanit, 2012) was due to the essential constituents in Jasmine such as linalool, fernesene, jasmonaic acid, phenylpropanoids, benzyl acetate and nerolidol (Pragadheesh et al., 2017). These components are very important in formulating deodorant in cosmetic field. However, these bioactive compounds do not last long and easily volatile therefore encapsulation of bioactive component has long been pursued in the search for extend shelf life in food, beverages and pharmaceutical industry (Teeka, Chaiyasat, & Chaiyasat, 2014).

Table 2 Common bioactive compounds in Jasmine flower

Compound	Molecular Structure	Solubility
Linalool		Hydrophobic

Jasmonic acid		Hydrophobic
Farnesene		Hydrophobic
Benzyl Acetate		Hydrophobic

2.3 Encapsulation with β-cyclodextrins

Encapsulation can be defined as a process of entrap one substance to another substance, thereby producing particles with diameters of nanometer to micrometer. Encapsulates can be distinguished into two types which are capsule and matrix type. The capsule type is also called reservoir, single-core, core-shell or mono-core type. The capsule type has an active agent being entrapped by a shell while for the matrix type, the active agents are present at the surface or surround encapsulates (Zuidam, 2010). Encapsulation of sensitive materials generally consisted of two steps, the first step is emulsification of core materials with a dense solution of the wall materials while the second step was drying or cooling of the emulsion (Madene et al., 2006). Encapsulation offers many advantages such as increase the physical stability of bioactive compound during the process (Fakhreddin et al., 2013), preserved the quantity of bioactive compound, and protect a fragile or unstable compound from the

adverse environmental effect during storage (Munin & Edwards-lévy, 2011). Encapsulation can reduce the rate of volatile and prevent degradation from heat, air and light (Risch, 1995).

Cyclodextrins (CD) are used as encapsulating agent due to their ability to encapsulate hydrophobic guests into their cavity through the formation of CD/guest inclusion complexes. This process was stabilized and involved a wide variety of intermolecular interaction, such as formation of hydrogen bonds, Van der Waals interaction and hydrophobic interaction. CD is cyclic oligosaccharides and can be represented as a truncated cone structure with hydrophilic external surface and hydrophobic cavity. The structure of CDs is stabilized by the formation of hydrogen bonds between C-2 and C-3 hydroxyl groups of adjacent glucose units (Kfoury, Hădărugă, Hădărugă, & Fourmentin, 2016). CD encapsulation of bioactive compounds has the ability to alter the physiochemical characteristics of both materials.

A study has shown that cyclodextrin (CD) have the ability to form inclusion complexation with functional food substances such as antioxidants, antibacterial, flavors and aromas thus can increase the stability, solubility, reactivity and sustained release of the encapsulated bioactives (Pinho et al., 2014). From a study, solid lipid nanoparticles based on stearic acid and ethyl cellulose were used to encapsulate maltol flavor without interaction between the ingredients and chemical. The stability of encapsulated nanoparticles (10-100 nm) had enhanced and increase resistance towards flavour loss or degradation during processing and storage (Eltayeb, Bakhshi, Stride, & Edirisinghe, 2013). Recent studies have shown that the ability of CD to form inclusion

complex with linalool. In this study, the bioactive compounds referred to linalool since it is the most abundant in flora scent which is up to 70%. Linalool is an acyclic monoterpene tertiary alcohol with hydrophobic nature, therefore inclusion complex of linalool into CD to overcome the drawbacks of linalool, including low solubility and stability (Aytac, Yildiz, Kayaci-Senirmak, Tekinay, & Uyar, 2017).

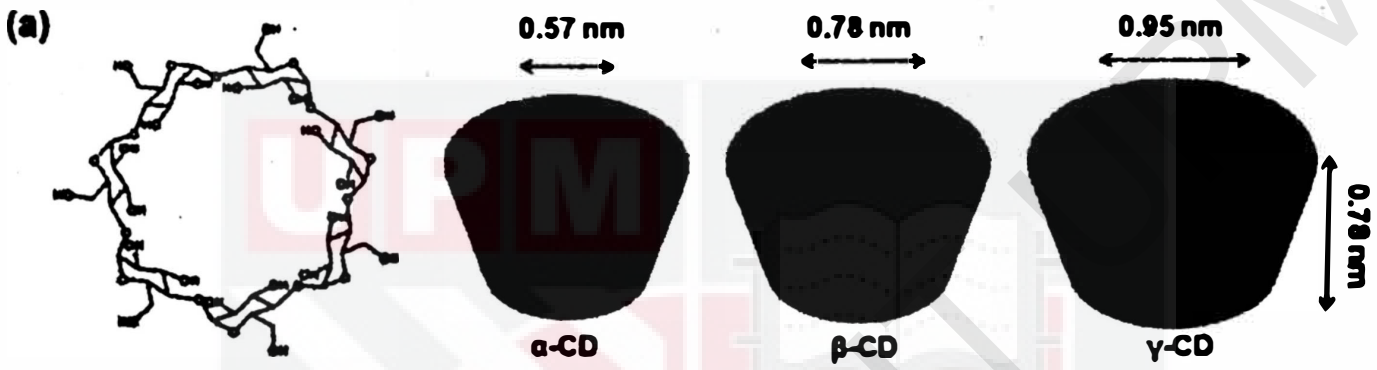


Figure 3 Schematic representation of CD (Kayaci, Ertas, & Uyar, 2013)

2.4 Electrostatic Atomisation

Electrospraying and electrospinning are the electrohydrodynamic processes where the solute solution can be sprayed or spun by the high electrostatic field to obtain particles or fibers, respectively. Electrostatic atomisation or electrospraying is a technique used to produce very fine or mono particle droplets due to the electric static force exerted on the surface of the produced droplet (Yurteri, Hartman, & Marijnissen, 2010). When the number of ions like charged increased at the tip of capillary, electrostatic repulsion and coulombic forces of external electric field occur thus distorted the hemisphere surface of droplet into conical shape that is known as Taylor Cone. When the suspension concentration is high, the jet will become destabilised due to varicose instability and form the fine droplets. The agglomeration and coagulation

can be avoided due to high density charged droplets are self-dispersing in space (Bhushani, 2014).

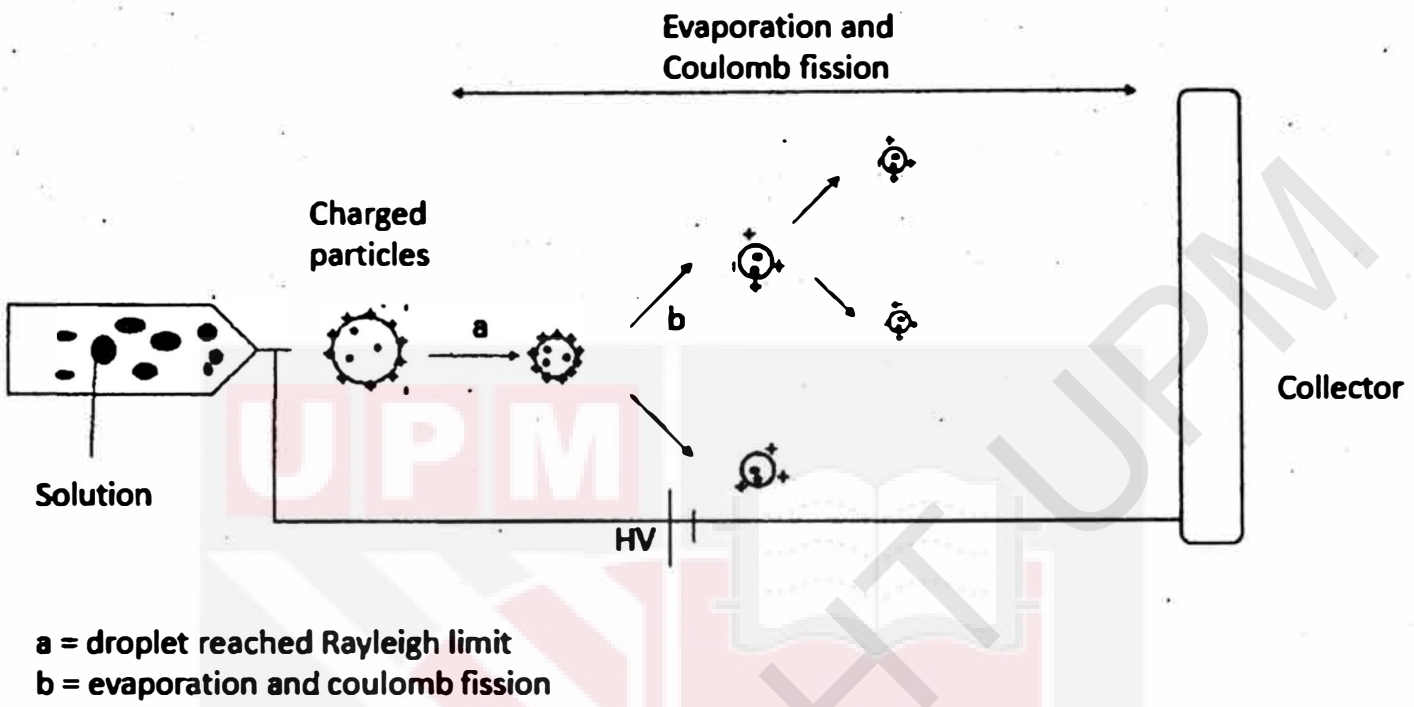


Figure 4 Schematic illustration of electrospray drying mechanism

2.4.1 Electrospray Parameter

The size of the solidified particles formed for the thin film coating can be controlled by adjusting the factors such as the system, solution, instrumental and ambient parameters. It was desired to get the small size of droplets because the large surface area of the nanoparticles increases its probability of agglomeration or aggregation thereby causing size growth. Firstly, the concentration of the suspension and solvent used that need to determine its properties which including pH, conductivity, viscosity and surface tension. Secondly, the instrumental parameters include voltage supplied, flow rate of the suspension, distance between the tip of the needle and the

collector and the nature of collector material (Jaworek & Sobczyk, 2008). Additionally, the ambient conditions such as the temperature, humidity and air velocity in the process chamber together determine the rate of evaporation of the solvent from the electrosprayed product.

a. Concentration and type of suspension and solvent used

A studied from Lopez (2012), for certain material the size of the capsule can be reduced when decreased the concentration of the suspension. In this case, whey protein concentrate (WPC) was tested with different concentration and results showed that high concentration which is more than 35% of suspensions were needed to obtain electrospray capsule. For concentrations below 35% only drops of material were collected. In contrast, a suspension with concentration more than 60% a gelled structure was collected from the unstable jetting during the electrohydrodynamic process. By varying the pH of the suspension, it leads to significant changes of capsule size where the optimum pH that can obtained the largest size is pH 6.4.

Based on a study of encapsulation of cisplatin in poly (lactic-co-glycolic acid) nanoparticles for controlled drug delivery, the FTIR analysis showed that a lower concentration of cisplatin in the encapsulating agent, no peaks corresponding to cisplatin were observed (Parhizkar et al., 2016).

b. Instrumental parameter

Electrospray aids in uniform coating of large areas of substrate with adequate control over the deposition rate and film thickness. Also, coating of submicron or nanoscale, monodisperse particles with narrow size distribution is required to reduce the number

and size of voids and cracks in the film. Recently, spray coating of chocolate by electrohydrodynamic spraying has been studied by researchers. Parameters such as viscosity, yield value and electrical resistivity of chocolate were found to have a significant effect on the spray quality. It has concluded that decreased in these values can decrease the size distribution of droplets formed and caused uniform coating (Bhushani, 2014).

c. Voltage

The voltage applied is important parameters as a driving force for the electrospray process. The voltage applied is used to overcome the surface tension of the suspension to form particles, thus high voltage is applied to deal with surface tension stress. According to Chakraborty (2009), when the electric field strength increases, the size of particle reduced significantly. However, when the strength of electrical field increased, it will lead to instability of spray mode.

d. .Other parameters

The other parameters that will affect the size and morphology of the particles are distances between the capillary needle and the collector, temperature and humidity. Temperature and relative humidity played an important role in produced fine and spherical electrospray particles. If the temperature is too high, the molecular mobility increases due to the energy available for evaporation of water from the suspension and increase the surface roughness of the products (Bringas-Lantigua, Valdés, & Pino, 2012). When the temperature increases, solution conductivity will increase due to the dissociation of ions and resulted in decrease of solution viscosity and surface tension

(Zone & Clare, n.d.). The evaporation rate of solvent can be decreased by increase the relative humidity in the electrospray process and resulted in larger particle diameters (Bhushani, 2014). The increase in distance between the needle and the collector lead to a smaller particles in size due to the longer distance allows enough time for evaporation to take place (Chakraborty, Liao, Adler, & Leong, 2009).

2.5 Drying mechanism of atomized droplets

Drying is a separation process by converting the solid, semi-solid or liquid into a solid product by means of evaporation of liquid into vapor phase. According to Parikh, drying is a complex unit operation that involved heat and mass transfer along the process which caused the physical and chemical transformation in the product. Drying take place when there is a vaporization of liquid by heat or without heat supply. For example, drying with and without heat is spray drying and freeze drying respectively. Spray drying is a unit operation by which the liquid droplets were atomized in a hot air chamber to produce a fine powder (Gharsallaoui, Roudaut, Chambin, Voilley, & Saurel, 2007). In freeze dying, products containing liquid are dried in the system while being maintained at a temperature below that of its crystallization point. Freeze drying is based on the principle of dehydration by sublimation of frozen product. This processing technology is suitable for heat sensitive materials since it is a non-thermal drying technique and exerts less stress on the materials (Librán, Castro, & Lagaron, 2017). However, one of the drawbacks of this technique is required higher cost for the equipment.

Typically, drying of droplets consisted of solids can be divided into two stages. In the first stage, the droplets containing liquid was entered into the drying space where evaporation occurs caused shrinkage of the droplet diameter. Secondly, drying process is continuous to occur when the droplets moisture content falls down to a critical value until a dry solid crust surrounding a wet core is formed (Taylor et al., 2010).

2.5.1 Water droplet evaporation in ambient condition

During electrospray atomization, the solvent was evaporated from the charged droplets formed, therefore it was possible to produce fine and dense particles that have uniform in size. The drying condition during the electrospray drying method will affect the properties of the produced particles, for example fast drying rate resulted in the formation of hollow and porous structure of particles due to short time for solvent evaporated from the atomized droplets (Nyström, Murtomaa, & Salonen, 2012). Solvent evaporation occurred at the charged droplets produced at the spray needle caused the shrinkage of the droplets while the charges remain constant. The repulsion between the charges at the surface of the droplets surface increases until it overcome the cohesive force of the surface tension at a certain droplet radius.

The availability of the moisture content in the charged droplets is depending on the surrounding humidity and temperature in which can relate to psychrometric chart (L.Earle, 1983). The psychrometric chart is a graphical representation of the psychrometric processes of air which give the information on the temperature (wet-bulb and dry-bulb), relative humidity, enthalpy and air density. Psychrometric chart is defined as the study of the relationship between the material and energy balance of

water vapour-air mixture. Psychrometric chart summarizes the important calculations on how much heat and moisture can be added to or removed from the air.

According to Parikh, for drying to occur, there must be a concentration gradient, which must present between the moist particle and surrounding. Relative humidity (RH) of surrounding will affect the drying capacity of the air. At 100% RH, the air possesses the maximum amount of water at a given temperature, when the temperature of air is increased, it will cause the drop in RH. Hence, the drying capacity will be varies and depends on the processing conditions.

In the electrospray drying, the evaporation of solvent from charges droplets involves complicated process of heat and mass transfer that caused the shrinkage of particles (Sano & Keey, 1982).

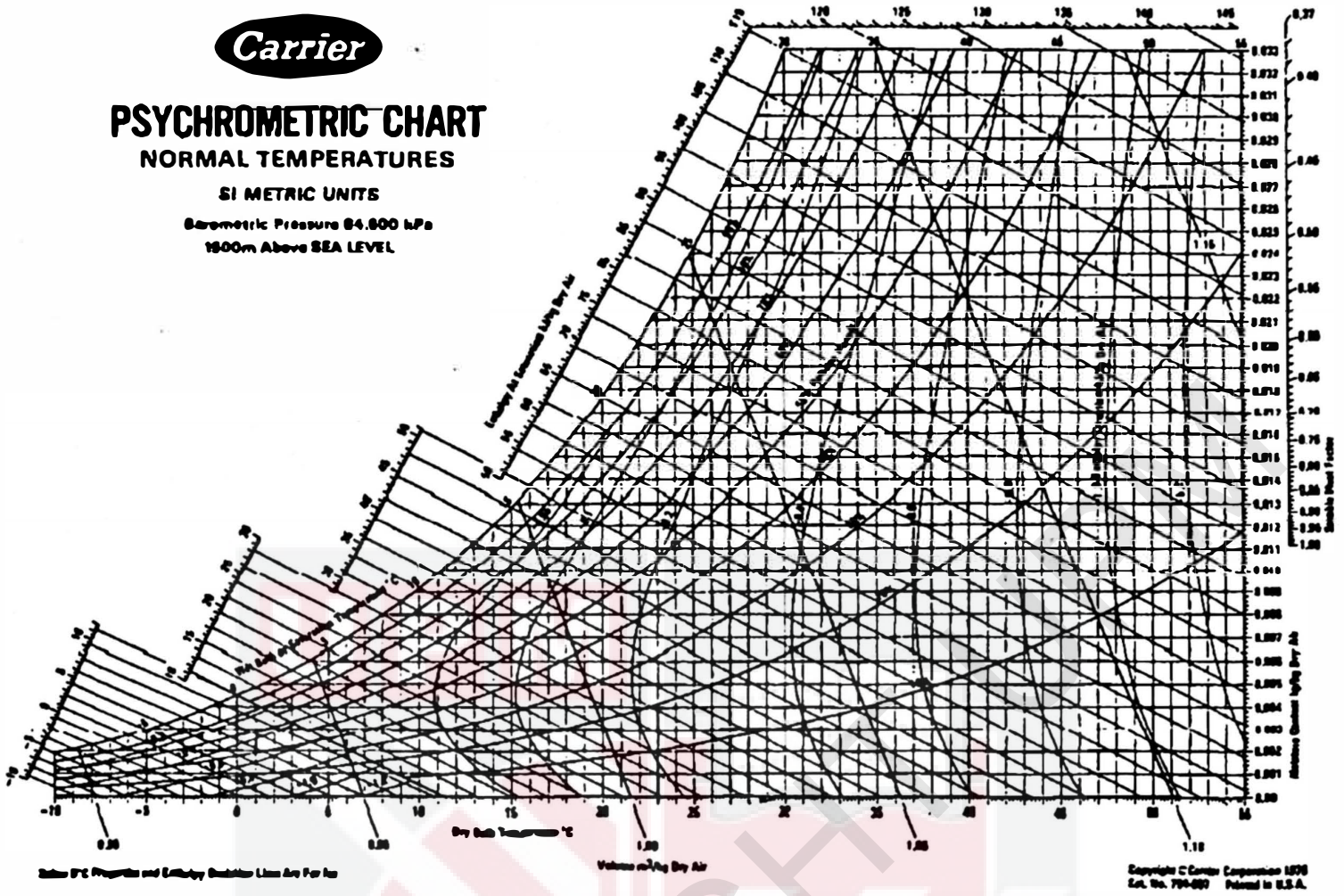


Figure 5 Psychrometric charts (L.Earle, 1983)

CHAPTER 3

METHODOLOGY

3.1 Jasmine Sample

Jasmine flowers were plucked in the area of eleventh college, University Putra Malaysia. Select the fresh Jasmine flowers that were in good conditions and without insects or worms. New samples were obtained to carry out every experiment.

3.2 Extraction of Fragrance Compound into Water

3g of fresh jasmine flowers which is approximately 25 samples flower were mixed with 60ml of water (ratio 1: 20) and undergo Ultrasound-Assisted-Extraction (Fisher Scientific Model FB 705, ½ inch probe) at 700 watts and 20 kHz for 1 hour. UAE caused the disruption of biological cell wall thus improve the performance in penetration of solvent and extraction. In this process, the beaker contained samples was immersed inside a container that was filled up with ice in order to absorb heat generated when the extraction process was carried out. After the extraction, the suspension was filtered using filter cloth to remove any impurities. Then the suspension was centrifuge at 10000 rpm for 30 minutes at 20°C. The clearer solution was then concentrated in a rotary evaporator at 60°C with 90 rpm until removal of the

solvent is complete. The materials that coated on the wall of conical flask was taken out using a spatula. The concentrated and dried solute was stored in the refrigerator at 4 °C for further preparation of different concentration of Jasmine solution which are 5 wt%, 15wt% and 25wt%.

3.3 Encapsulation

For the preparation of samples with encapsulated with β-cyclodextrin, different concentration of Jasmine solution will then added with β-cyclodextrin (CD) in a ratio of 10:1(w/v) and stirred by using magnetic stirrer for 2 hours. CD was added in small quantity from time to time to ensure uniform coating of Jasmine solution.

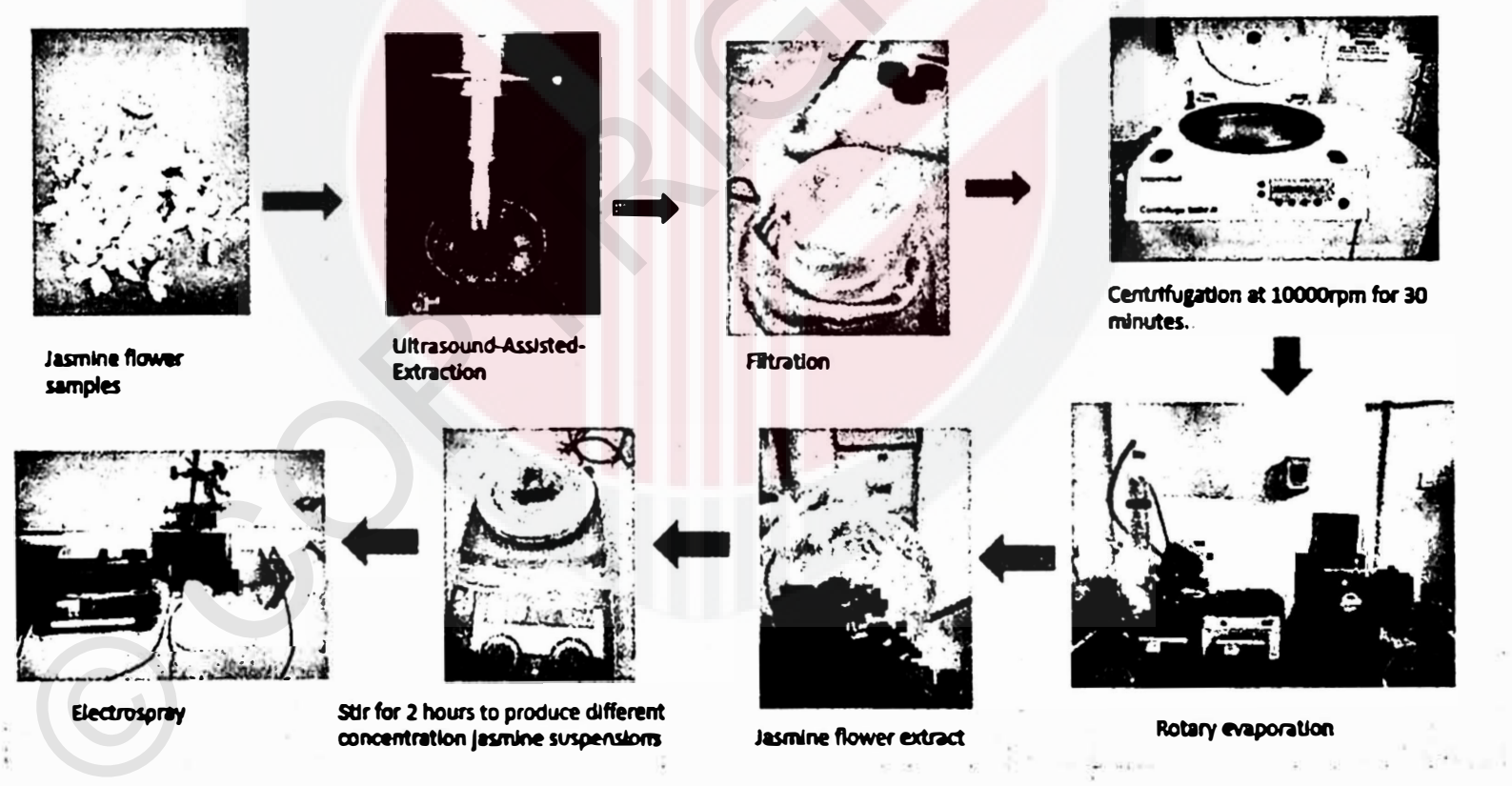


Figure 6 Steps for extraction of jasmine flower

3.4 Electric Assisted- Atomization (Electrospray)

3.4.1 Setup of Electrospray System

The electrospray system required few basic elements such as power supply, syringe pump, syringe nozzle, electrode ring and a collector attached with aluminium foil. The syringe contained jasmine flower solution that connected with syringe needle which act as a nozzle was placed on the syringe pump. The collector was placed opposite of the nozzle with a distance of 30cm from the syringe nozzle which is the optimum distance to get small particles (Parhizkar et al., 2016). The ring electrode was placed perpendicular to the syringe pump to stabilize the atomized droplets formed (Yurteri et al., 2010). The positive charge supply was connected to the nozzle while the ring and aluminium foil attached with the collector plate were grounded and the circuit was complete.

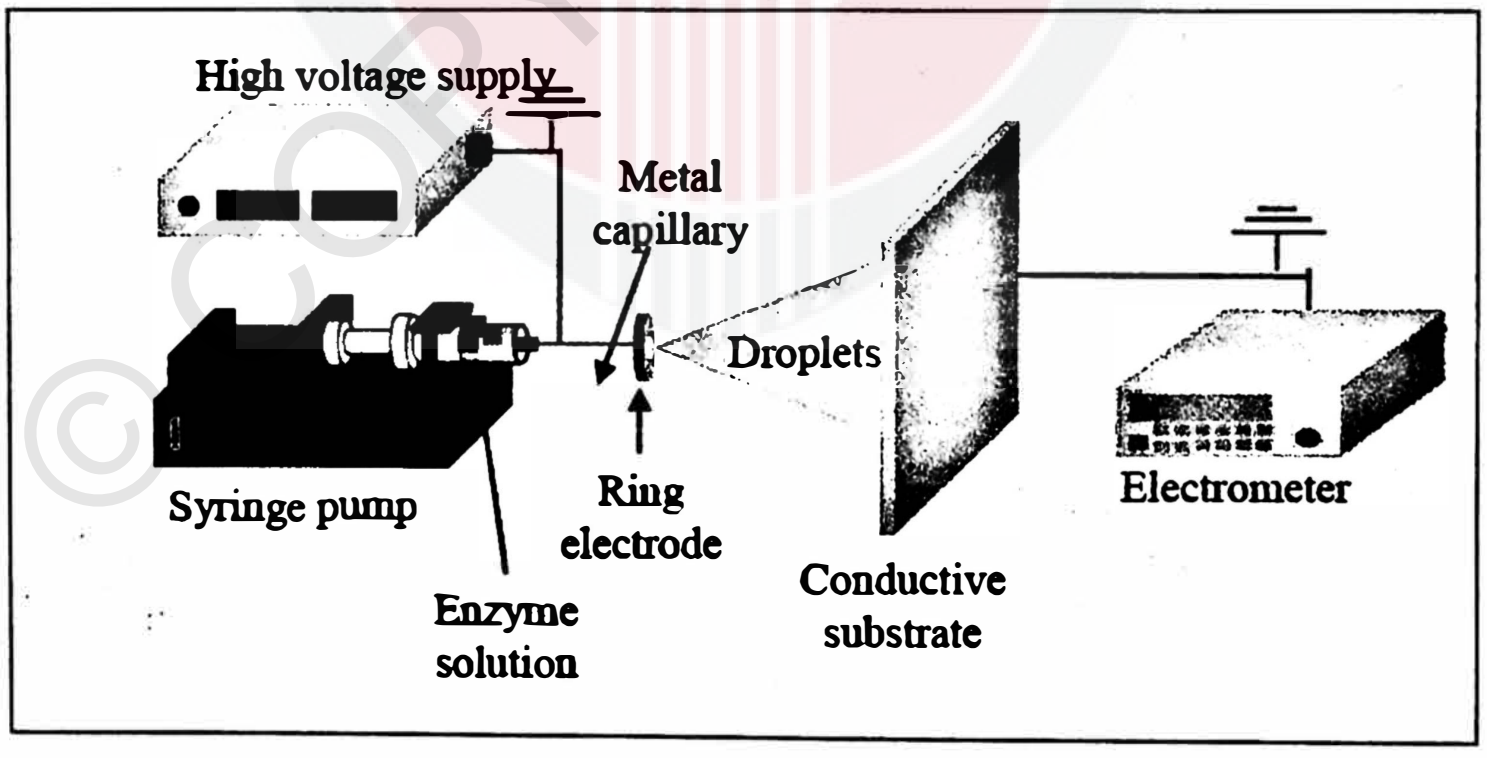


Figure 7 Electrospray-set up

3.4.2 Electrospray parameters

There are several parameters that played an important role in order to obtain a very stable electrospray condition such as voltage supplier, flow rate of the solution, the geometry of system used and the dielectric strength of the ambient medium. The properties of solution including surface tension, electrical conductivity, viscosity and density (Chakraborty et al., 2009). In addition, the humidity and temperature of surrounding of the room might affect the formation of stable Taylor-cone mode that caused the formation of mono-dispersed nanoparticles. Thus, it should be constant at temperature of 18 - 22°C and at enclosed room.

Experiment was carried out in trial and error approach in order to get the stable cone-jet mode by manipulating the voltage supplied and also the distance of nozzle and substrate (collecting foil). Firstly, the suspension from the capillary nozzle was in dripping mode, after manipulating the voltage supplied, the Taylor cone formed from the nozzle when the interface between the conducting liquid and air is electrically charged beyond the critical level. Once the stable Taylor cone was established, the collection of particles was ready and undergo electrospray for 6 hours.

Table 3 Operating parameters of electrospray

Parameter	Range
Voltage (kV)	4 – 5
Flow rate (ml/hr)	0.2
Needle tip to substrate distance (cm)	30
Electrospray duration (hr)	6

3.5 Characterization of Results

3.5.1 Field Emission Scanning Electron Microscopy (FESEM)

The morphology and electrosprayed particles were studied using Field Emission Scanning Electron Microscopy (FESEM). FESEM samples were prepared by collecting electrosprayed particles on aluminium foil. Before the analysis, the samples were sputter-coated with platinum under vacuum to avoid charging effect during the analysis. High magnification of the collected samples were carried out at Institute of Advance Technology (ITMA) of UPM. The model of equipment used to analyse the enlarge image of the samples is NOVA NANOSEM 230.

3.5.2 Fourier Transform Infrared Spectroscopy (FTIR)

Fourier Transform Infrared Spectroscopy is used to analyse the collected particles on the collector by interpret the peaks of wavelength and transmission to identify the interaction between the components. It is a technique used to obtain an infrared spectrum of absorption, emission of a solid, liquid or gas. The main purpose of this analysis is to demonstrate the interaction between the CD and the jasmine extract. FTIR spectra will indicate the additional peaks or absence of the characteristics peaks corresponding to the bioactive compound and encapsulating agent.

The samples collected on the aluminium foil were analyzed and pattern on graph were compared between the first week and the after one week of storage. FTIR of the collected samples were carried out at Institute of Advance Technology (ITMA)

of UPM. The model of equipment used to analyse the FTIR spectra is Thermo Nicolet 6700 and the spectra range is set from 400 to 4000 cm^{-1}.

3.5.3 Thermo gravimetric analysis (TGA)

TGA able to test the thermal stability and properties of encapsulated and non-encapsulated jasmine flower extract. The weight losses were observed in the TGA curves where these weight losses were attributed to the water loss and main thermal degradation of cyclodextrins (CD), respectively (Aytac et al., 2017). TGA measurements were performed using Mettle Toledo model TGA 1 HT at Institute of Advance Technology (ITMA) of UPM and the samples were heated from 25 °C to 400 °C at the rate of 10 °C/min.

3.5.4 Stability test

The electrosprayed nanoparticles were stored in the desiccator with silica gel in the laboratory for a certain period of time. After 28 days of storage, small amount of samples was analysed by TGA in order to determine the shelf life and thermal stability of samples. The samples were also analysed by FTIR after 28 days of their storage to investigate the existence of bioactive compounds in the electrosprayed nanoparticles.

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Rate of water removal

Electrospray technique involved the mechanism of water removal from the droplet along the trajectory path towards the collecting plate. In this study, the rate of water removed from the initial wet jasmine extract droplet to the final particle size was investigated by using the drying rate equation (L.Earle, 1983):

$$\frac{dw}{dt} = kA\Delta Y \qquad (1)$$

Where

$\frac{dw}{dt}$ is the mass being transferred (kg/s);

k is the mass transfer coefficient (kg/m^2s);

A is the area through which the transfer is taking place (m^2);

ΔY is the humidity difference (kg/kg)

The experiment is conducted at a constant temperature of 20°C and at varies relative humidity (RH). The amount of moisture removed (ΔY) can be obtained from the psychrometric chart based on the difference in RH and temperature. Based on the

equation, the relationship between the drying rate and the area of the spherical particle which the transfer is taking place and moisture removed based on the difference in relative humidity was identified.

The rate of water removed was tabulated in the table 4 below:

Table 4 Data of drying rate curve

	Distances	$\Delta V(nm^3$ x $10^8)$	t (s)	dw/dt (10^{-13}kg/s)	$A(nm^2)$	ΔY(kg water/kg product)	$A\Delta Y$ (nm^2kg/kg)
5wt%	10	2.6	0.12	2.17	266948.8	0.0029	774.1516
	20	3.71	0.23	1.61	104062.4	0.004	416.2494
	30	3.94	0.35	1.12	18869.24	0.0051	96.2331
15wt%	10	2.28	0.11	2.07	188574.5	0.0031	584.5811
	20	3.26	0.22	1.48	90365.49	0.0041	370.4985
	30	3.47	0.32	1.08	13467.96	0.0056	75.42059
25wt%	10	1.96	0.1	1.96	152260.9	0.0032	487.2347
	20	2.79	0.19	1.46	76258.07	0.0046	350.7871
	30	2.98	0.29	1.02	9837.109	0.0062	60.99008

From the figure 8, the drying rate of solidified jasmine was identified which is increasing for each concentration (5wt%, 15wt% and 25wt %) due to the change in humidity increased, thus it facilitates the water removed from the solidified jasmine extract. In this case, the relative humidity plays an important role in rate of evaporation when deal with small volume droplets thus will affect the deposition of the droplets (Brutin, 2013). Theoretically the rate of drying will increase when the relative humidity is low and decreased when the relative humidity is high. In order to verify the application of the drying rate equation when the humidity difference was increased, the approach of 'radius-square-law' was demonstrated which is proposed by Wang, Yao, Yang, Liu, and Jin, (2017) and applicable for the isothermal condition where no surface tension effect and ΔT nearly approximately to constant.

$$d_0^2 - d_p^2 = kt \qquad (2)$$

where d_0^2 is initial droplet diameter, d_p^2 is the diameter of particle deposited at any time t, and k is the evaporation rate constant.

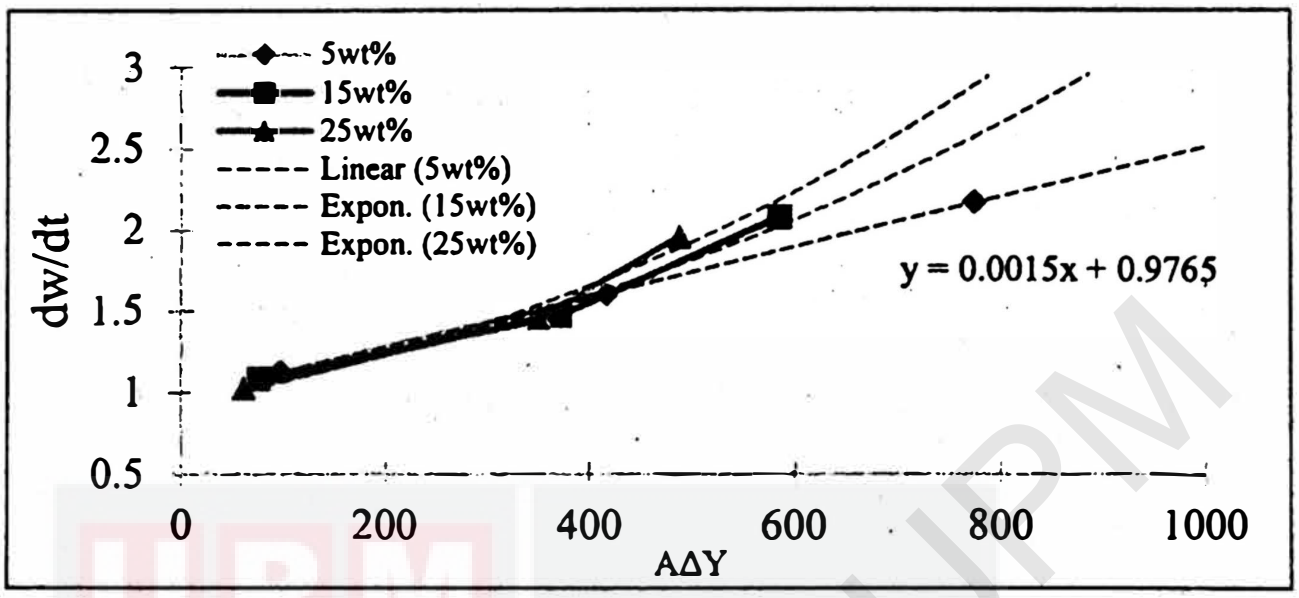
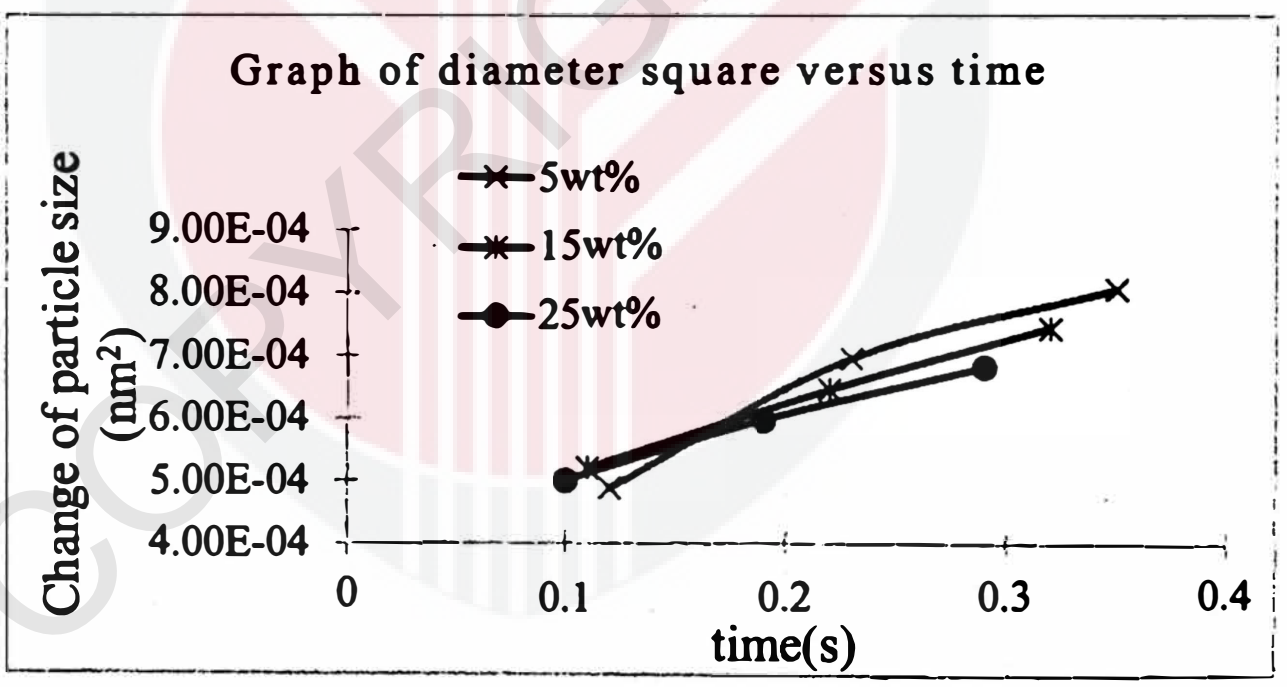
$$d\,(d^2) = kt \qquad (3)$$

According to the table 4, the relationship between the mass transfer coefficients in equation 1 and the evaporation rate constant in equation 2 was identified based on the graphs plotted. Both of the k values are related to the rate of water removed, therefore, based on the graphs plotted, hereby can deduced that both equations depicted the similar trend even though two different drying mechanisms. For the drying rate curve, the water removed increased when the difference in the humidity increased while in the other case, the difference in particle size increased when the time increased for each different concentration of jasmine extract.

Theoretically, the effect of humidity is proportional to the drying rate, however, in this case study, at fixed distance of 30cm, the rate of drying decreased with the increased of humidity difference when the concentration increased from 5wt% to 25wt% which is $1.12x10^{-13}$ kg/s and $1.02x10^{-13}$ kg/s respectively. This phenomena is most probably due to the crystalline bridges formed when the liquid evaporates. When the evaporation occurred, the proportion of water in the solidified particles decreased and produced a high strength powder, hence it is more difficult for water to be removed. In other words, electrostatic forces decreased rapidly when the humidity of surrounding air is increased from 0.0051 to 0.0062 kg water vapour/kg dry air for concentration of jasmine powder 5wt% and 25wt% respectively while this is due to the formation of liquid bridges with the moisture of the atmosphere, thus delay the evaporation process (Rhodes, 2008). Based on the drying rate curve, higher k value were noticed with more concentrated droplet which is 25wt% jasmine extract. The point whereby the dw/dt start to diverge is hypothesized causes by droplet fission phenomena. In electrohydrodynamic process, surface tension and conductivity of solution play an important role. When there is an increased of jasmine extract concentration, it will caused the increase of molecular weight, thus affect the viscosity, conductivity and surface tension of electrospray particle (Bhushani et al., 2017). The higher concentration contribute to the charge per mass ratio which lead to the increase of the electric field on the surface of the charged droplets as the droplets gets smaller (Cole, 2000). Finally, the repulsion between the charges at the surface of droplets increased and facilitate the Coulomb fission to occur (Kebarle & Verkerk, 2010).

On the other hand, based on figure 9, it was showed that the rate of evaporation for 25wt% is lower compared to the 5wt% due to the water proportion content in the solidified powder is decreasing when the process of evaporation taking place. The differences in the particle size diameters was decreasing from 8.06 $x10^{-4}$ nm^2 to 6.83 $x10^{-4}$ nm^2 for the concentration of jasmine powder content 5wt% and 25wt% respectively. This is due to the water proportion in the solidified powder in 5wt% is higher than that 25wt% of jasmine powder.

Table 5 Comparison of drying rate curve and evaporation curve

$\frac{dw}{dt} = kA\Delta Y$	
	Figure 8 Drying rate curve
$d(d^2) = kt$	
	Figure 9 Evaporation rate curve

4.2 Morphology of nanoparticles

Scanning Electron Microscope (SEM) imaging provides morphology of the captured droplet with the particles. Figure 10 showed the image of electrospray drying for non-encapsulated and encapsulated jasmine extract with CD. During electrospray drying, the evaporation of solvent from the charged nanoparticles involved the complicated heat and mass transfer that lead to shrinkage of particle to occur. Hence, the morphology of electrosprayed particles at different concentration and distances were studies. In this study, the concentration of the bioactive compound and CD was found no influence in the morphology of the solidified particle but affect the particle size (Gómez-Mascaraque, Casagrande Sipoli, de La Torre, & López-Rubio, 2017). Therefore, the maximum concentration tested 25 wt% of the non-encapsulated and encapsulated jasmine extract was selected for further experiments in order to maximize the bioactive compound in the encapsulated structures. The morphology of the encapsulated particle in the detailed SEM image revealed the sphericity and smooth outer surface compare to the non-encapsulated particle. On the other hand, the morphology and deposition of non-encapsulated particles can be observed in the SEM image which agglomeration in contact with each other. The agglomeration of particles is due to the high water content that not fully evaporate, therefore the particles are not discrete (Yao, Kuang, Xie, Hua, & Wang, 2008). According to Nandiyanto and Okuyama (2011), the most stable shape for a droplet is spherical shape, thus the encapsulated jasmine extract possess higher stability compare to the non-encapsulated jasmine extract.

(a) (b)

Figure 10: FE-SEM images for non-encapsulated (a) and encapsulated particle (b)

4.3 Efficiency of Encapsulation

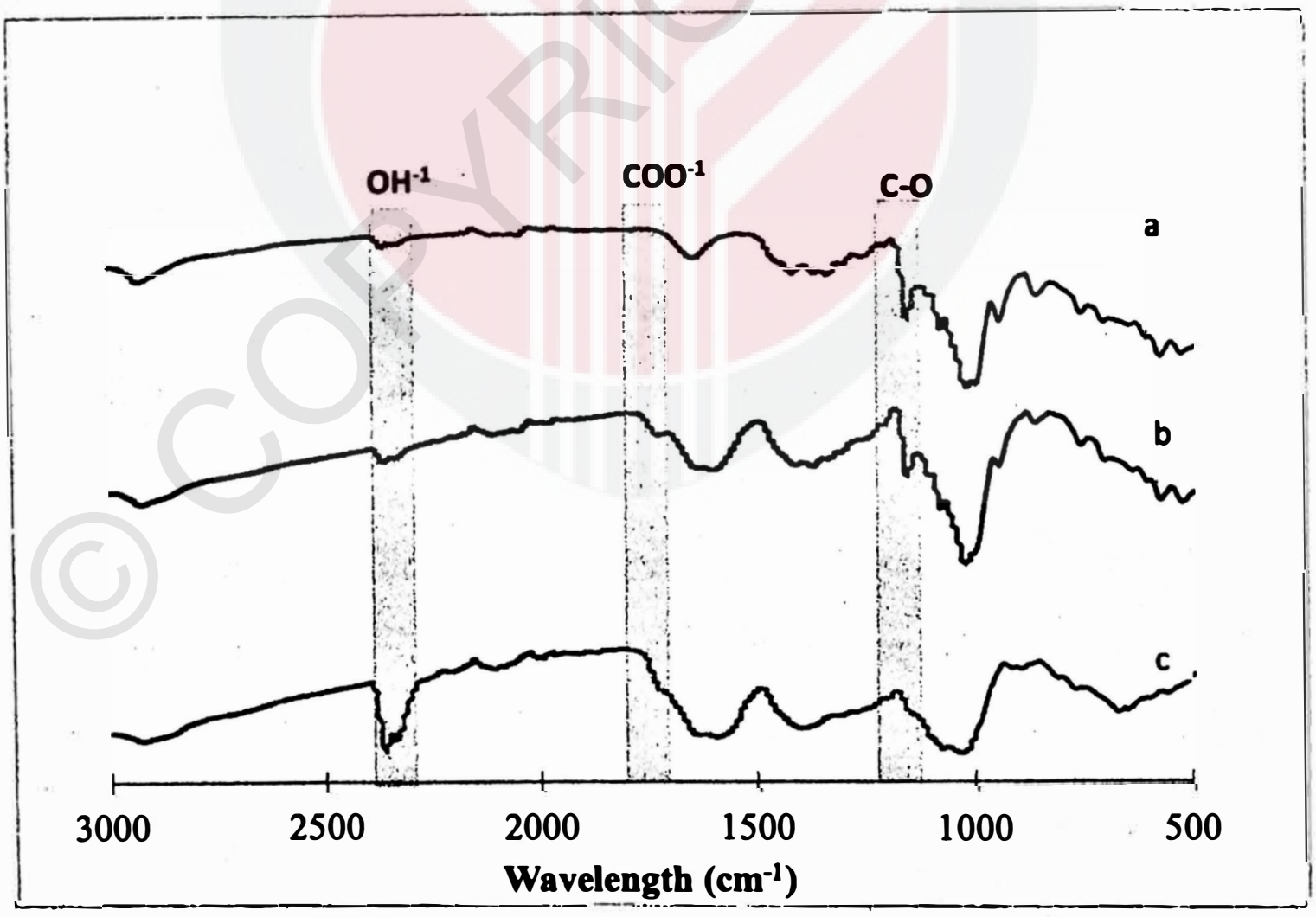


Figure 11 FTIR spectra of (a) β-CD, (b) encapsulated of jasmine compound and (c) non-encapsulated jasmine

FTIR is a very useful technique to prove the presence of both guest and host molecules in the inclusion complexation process. Figure 11 shows the FTIR spectra of the (a) β-CD, (b) encapsulated of jasmine compound and (c) non-encapsulated jasmine compound. The spectrum of β-CD look almost similar to the encapsulated jasmine compound which indicates the formation of inclusion complex between component β-CD and jasmine compound. Besides that, a broad hydroxyl band of pure CD was observed at 3264 cm^{-1}. The frequencies of CD observed at 3264cm^{-1}, 2926cm^{-1}, 1151 cm^{-1} and 1017 cm^{-1} which corresponds to the stretching of O-H, C-H asymmetric stretching vibration, C-C and C-O stretching vibration of the secondary alcohol groups that are present in the β-CD molecule respectively (Kotronia et al., 2017). Meanwhile, there is a very strong absorption band at 3273 cm^{-1} for O-H stretching vibration of water, 2924 cm^{-1} for C-H stretching vibration , 1029 cm^{-1} for bending vibration of O-H and lastly, 1593 cm^{-1} for C=C peak that present in the IR spectra of non-encapsulated jasmine extract which is also significant in the IR spectrum of encapsulated compound.

According to the spectra of infrared (IR) of encapsulated compound, the characteristics peaks attributable to stretching of O-H (3259 cm^{-1}), C-H stretching vibration (2923 cm^{-1}), C-C (1151 cm^{-1}), stretching vibration of C-O (1020 cm^{-1}) and C=C (1600cm^{-1}) which showed almost similar peaks with CD and non-encapsulated compound respectively. The appearance of the peak at 1409cm^{-1} most probably contained a combination of overlapped and shifting of the peak at 1414 cm^{-1} of the CD spectrum and 1397cm^{-1} of the non-encapsulated spectrum, which along with the characteristics absorption of new band that believe is belong to –COO group that exists at 1725cm^{-1} (HU Jing , XIAO Zuobing , ZHOU Rujun & Zhen, 2011) prove the encapsulation of

bioactive compound is take place, by indicating the presence of host-guest interaction (Kotronia et al., 2017). The shifting of the bands suggesting the formation of hydrogen bonds between the carbonyl groups of carboxylic groups and the hydroxyl groups of the host during inclusion complexes (Mura, 2015). Other than that, three of the FTIR spectra appeared the O-H stretching at the range of wavelength 2300 to 2400cm^{-1} which is belong to the moisture content in the samples CD, encapsulated and non-encapsulated Jasmine's extract. (Krumins, Klavins, & Seglins, 2012).

Table 6 and 7 showed the difference in frequencies between CD and encapsulated compound; and between non-encapsulated compound and encapsulated compound respectively.

Table 6 Comparison between the wavelength of CD and encapsulated compound

Functional Group	Wavelength (cm^{-1})	
	CD	Encapsulated Compound
[OH]	3264	3259
[CH_2]	2926	2923
[CC]	1151	1151
[CO]	1017	1020

Table 7 Comparison between the wavelength of Non-encapsulated and encapsulated compound

Functional Group	Wavelength (cm^{-1})	
	Non-encapsulated	Encapsulated Compound
[OH]	3273	3259
[CH_2]	2924	2923
[CO]	1029	1020
[C=C]	1593	1600

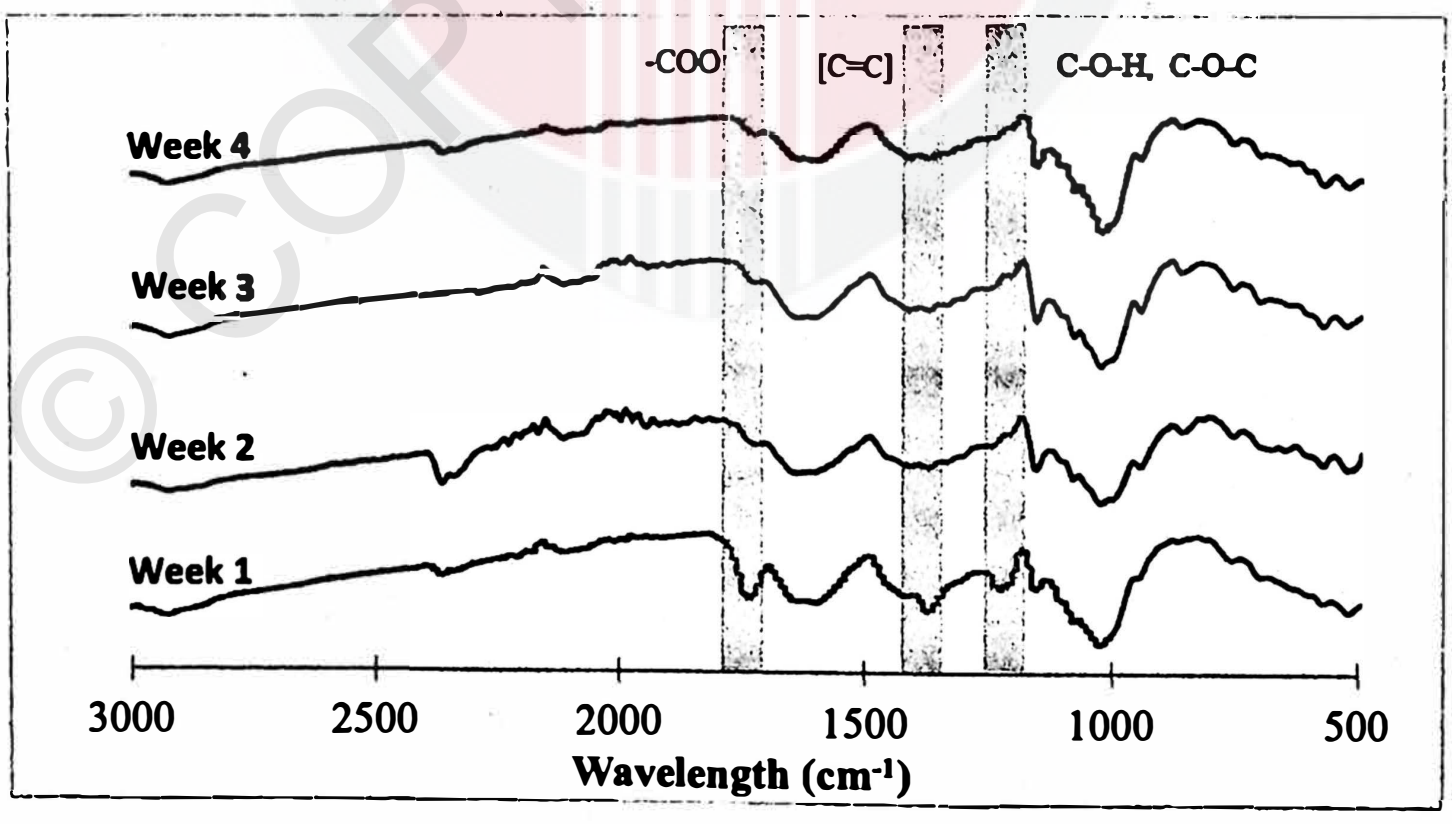


Figure 12 Comparison of FTIR spectra for encapsulated compound before and after storage

Figure 12 showed the differences of the FTIR spectra of the initial encapsulated compound (IEC) and the encapsulated compound after 28 days of storage (FEC). There is not much changes in the intensity and the absorbance of frequency for both IEC and FEC. The IR peak of IEC is obvious in the wavelength range from 1400-1600 cm^{-1} (C=C), but after their storage, the absorption band is not significant which confirmed the chemical interaction is not stable and remarkably weakened. Other than that, the IR peak at the range of 1200-900 cm^{-1} indicated the presence of coupling C-C, C-O stretching and C-O-H, C-O-C deformation modes of various oligosaccharides (Grube, Bekers, Upite, & Kaminska, 2002). The decreasing in peak ratio at this spectral region showed the effect of storage to these encapsulated compounds. The IR peak of the IEC was intense compare with the FEC at the wavelength range of 1900 - 1700 cm^{-1} which indicated the amount of new band stretching bonds (-COO) are depleting in the inclusion complex compound and relatively altered for subsequent days of storage (Man, 2011).

4.4 Thermal stability of nanoparticle

In this study, encapsulation of bioactive compound in beta-cyclodextrin (CD) inclusion complex nanoparticles was achieved via electrosprayed technique. The shelf life and thermal stability of the encapsulated bioactive compounds (EBC) was determined via thermogravimetric analysis (TGA). The electrosprayed EBC weas stored in the desiccator with silica gel to prevent any reaction with water for moisture sensitive materials. TGA is a useful technique in order to study the thermal stability and degradation of the fragrances after storage. The TGA studies were also performed

for the non-encapsulated bioactive compound (NBC) and EBC nanoparticles for comparison. Three steps of weight losses were observed in TGA curves for EBC and these weight losses were attributed to the water loss and the main thermal degradation of CD, respectively (Aytac et al., 2017). From the thermogram of NBC curve, the initial weight loss occurred at around 50°C corresponded to water loss and the second weight loss of bioactive compounds was started at 150°C. The bioactive compounds were further evaporated until 185°C when the temperature increasing which confirms it has volatile nature.

On the other hand, higher thermal stability of bioactive compounds was observed in the EBC TGA curve. TGA thermograms showed three weight losses, the first weight losses started at below 100°C due to water loss, the second weight loss was occurred between 110 °C to 200°C is due to the evaporation of bioactive compounds and finally, the major weight loss started at above 300°C corresponds to the main degradation of CD. Based on the Figure 13 the evaporation temperature of bioactive compounds shifted to higher temperature in the CD inclusion complex compared to the NBC which is from 150 °C to 190°C was due to the interaction with CD cavity (Kayaci et al., 2013). Thus, it can be deduced that cemplexation of CD with bioactive compounds was successfully occurred due to the shift in the thermal evaporation of bioactive compound to higher temperature (Aytac et al., 2017).

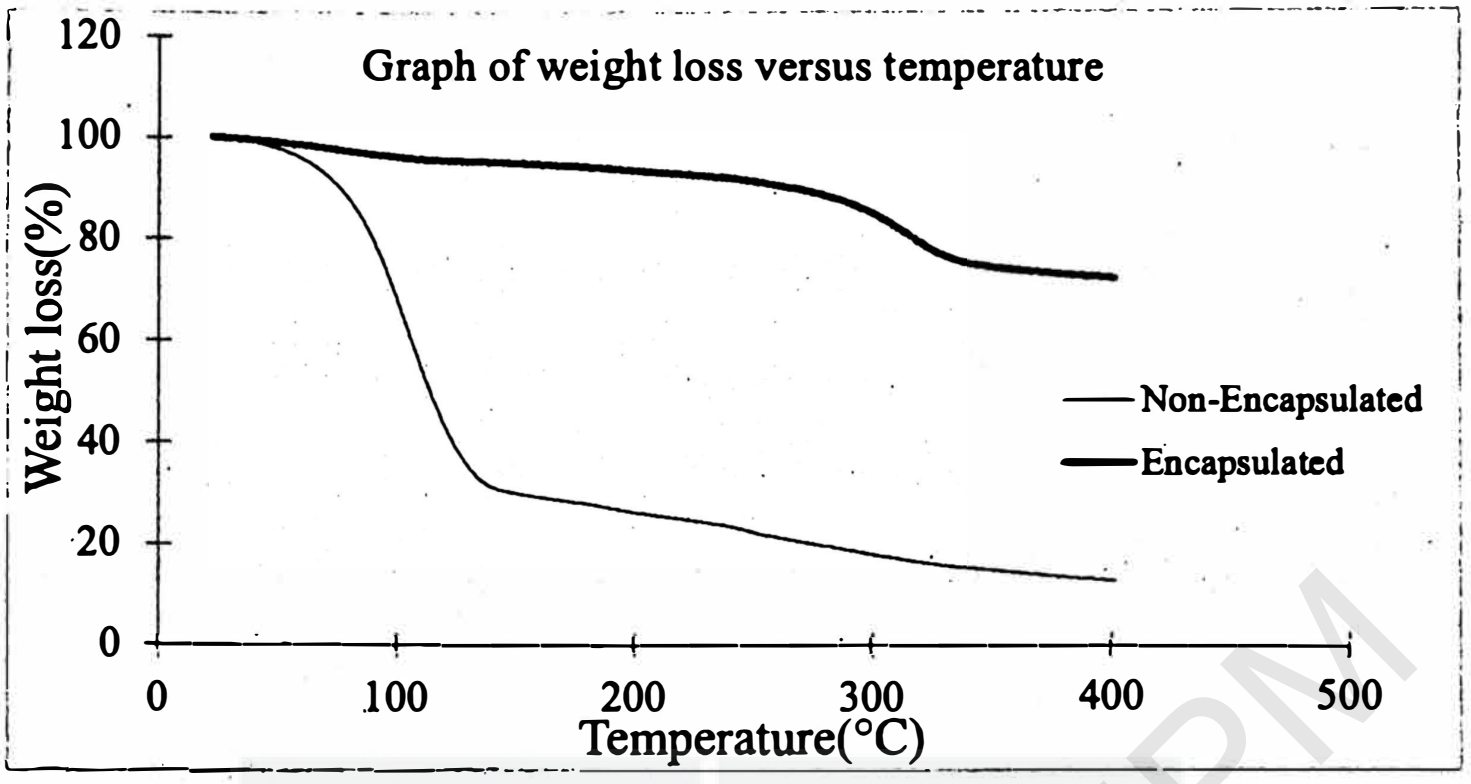


Figure 13 Thermogravimetric analysis (TGA) of the encapsulated and non-encapsulated sample

4.5 Loading Capacity of the bioactive compound

Figure 14 and 15 showed the degradation of the EBC after 28 days of storage in desiccator. Thermogravimetric curves of encapsulated bioactive compounds show a similar shape (Figure 14) except small differences in the zone of decomposition temperature and in the kinetics of the process. Basically, there were no significant changes in temperature shifting when there is a comparison between the first week sample and the samples after certain storage periods. Based on the figure 15 which is a plot of differential in weight loss versus temperature (DTGA) curve, the maximum decomposition of the sample is occurred at the range of temperature approximately 50-100°C, 180-220°C and 250-350°C. However the changes in percentage weight loss were decreasing after storage of 28 days where the area under the derivative of thermogravimetric curve getting smaller, this was due to the effect of storage, thus lead to the loss of the bioactive compounds for the encapsulated bioactive compound (Djaoudene & Louaileche, 2016).

The results are confirmed by calculating the loading capacity of the bioactive compound that are protected by CD in the encapsulated samples according to the equation 4 (Sansukcharearnpon & Wanichwecharungruang, 2010). Based on the table 8, the loading capacity of the sample in the first week is the highest which is 27.86% compared to the sample after storage of 28 days which is 22.68%. Hence, the results confirmed the effect of storage on the stability and the shelf life of the bioactive compound even though it has been encapsulated by CD.

$$\text{Loading capacity} = \frac{\text{mass of the entrapped compound(mg)}}{\textit{Initial mass of the sample(mg)}} \; x\; 100\% \qquad (4)$$

Table 8 Loading capacity of the encapsulated bioactive compound after storage of 28 days

	Initial Weight, M_o (mg)	Final weight, M (mg)	ΔM(mg)	Loading capacity (%)
Week 1	14.6788	10.59	4.0888	27.86
Week 2	10.4988	7.73	2.7688	26.37
Week 3	10.1509	7.82	2.3309	22.96
Week 4	7.6959	5.95	1.7459	22.68

Table 9 Loading capacity of the non- encapsulated bioactive compound after storage of 28 days

	Initial Weight, M_o (mg)	Final weight, M (mg)	ΔM(mg)	Loading capacity (%)
Week 1	10.1428	8.0593	2.0835	20.54
Week 2	11.7328	9.58	2.1528	18.35

Week 3	10.5409	8.67	1.8709	17.75
Week 4	12.9299	10.8860	2.0439	15.81

On the other hand, based on the figure 16 which is a DTGA curve of non-encapsulated jasmine compound depicted the changes in percentage weight loss were decreasing after storage of 28 days where the area under the derivative of thermogravimetric curve getting smaller. There is an obvious decomposition occurred at the range of temperature 25 to 130 °C and 150 to 230 °C due to the presence of water and bioactive compounds respectively. The results is supported by calculating the loading capacity of the bioactive compound content in the sample. Based on the table 9, it showed a significant reduced in loading capacity in the non-encapsulated bioactive compound which is from 20.54% decreased to 15.81% for the before and after storage of 28 days.

Thus, based on the Figure 13 and comparing the loading capacity of the bioactive compounds in encapsulated and non-encapsulated samples, we can deduce that the bioactive compounds can be protected using CD as encapsulating agent via electrospray technique, however, due to the effect of storage, it will lead to the degradation of bioactive compounds by calculating the loading capacity in the encapsulated samples.

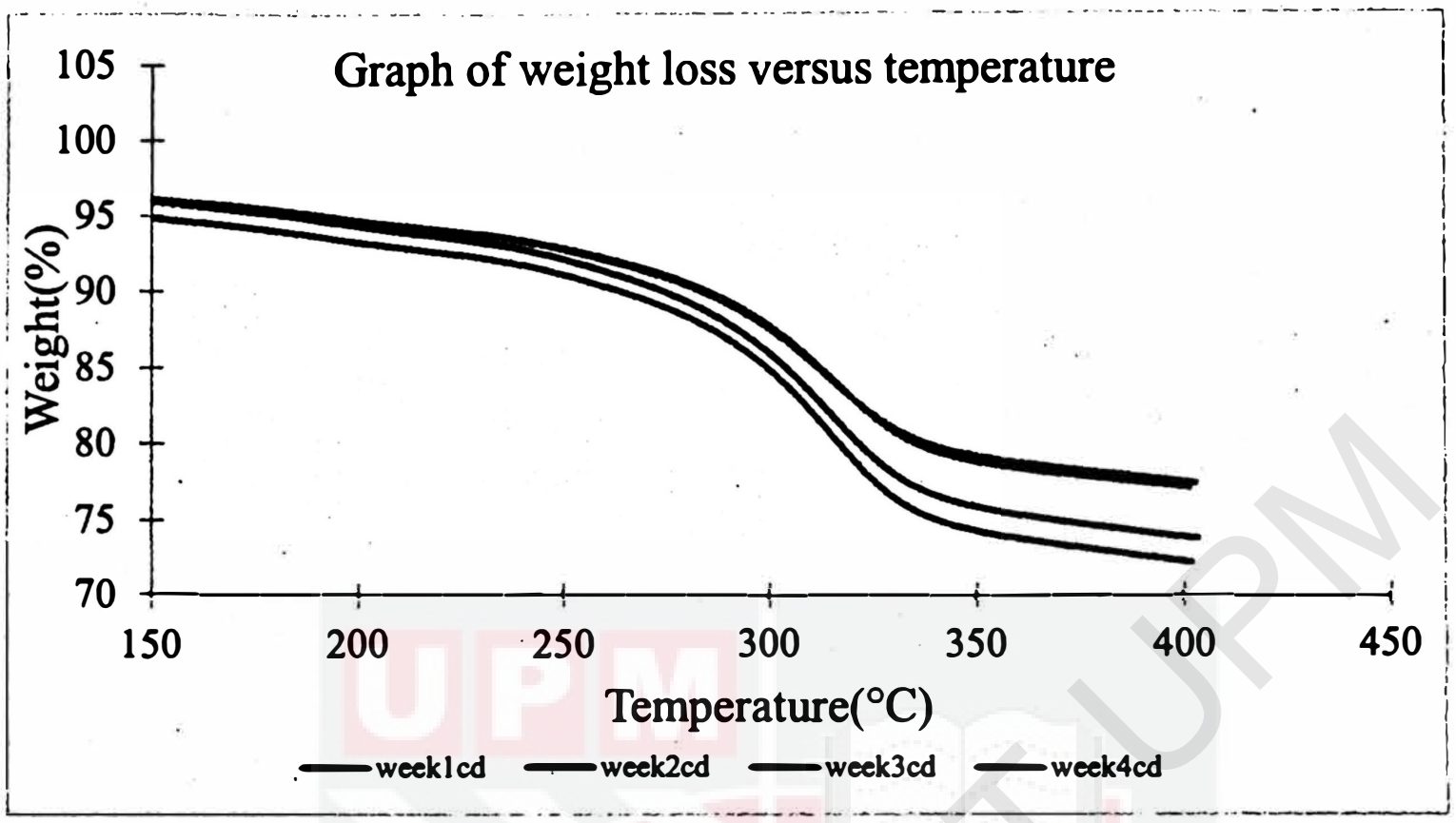


Figure 14 TGA curve of encapsulated samples after storage of 28 days

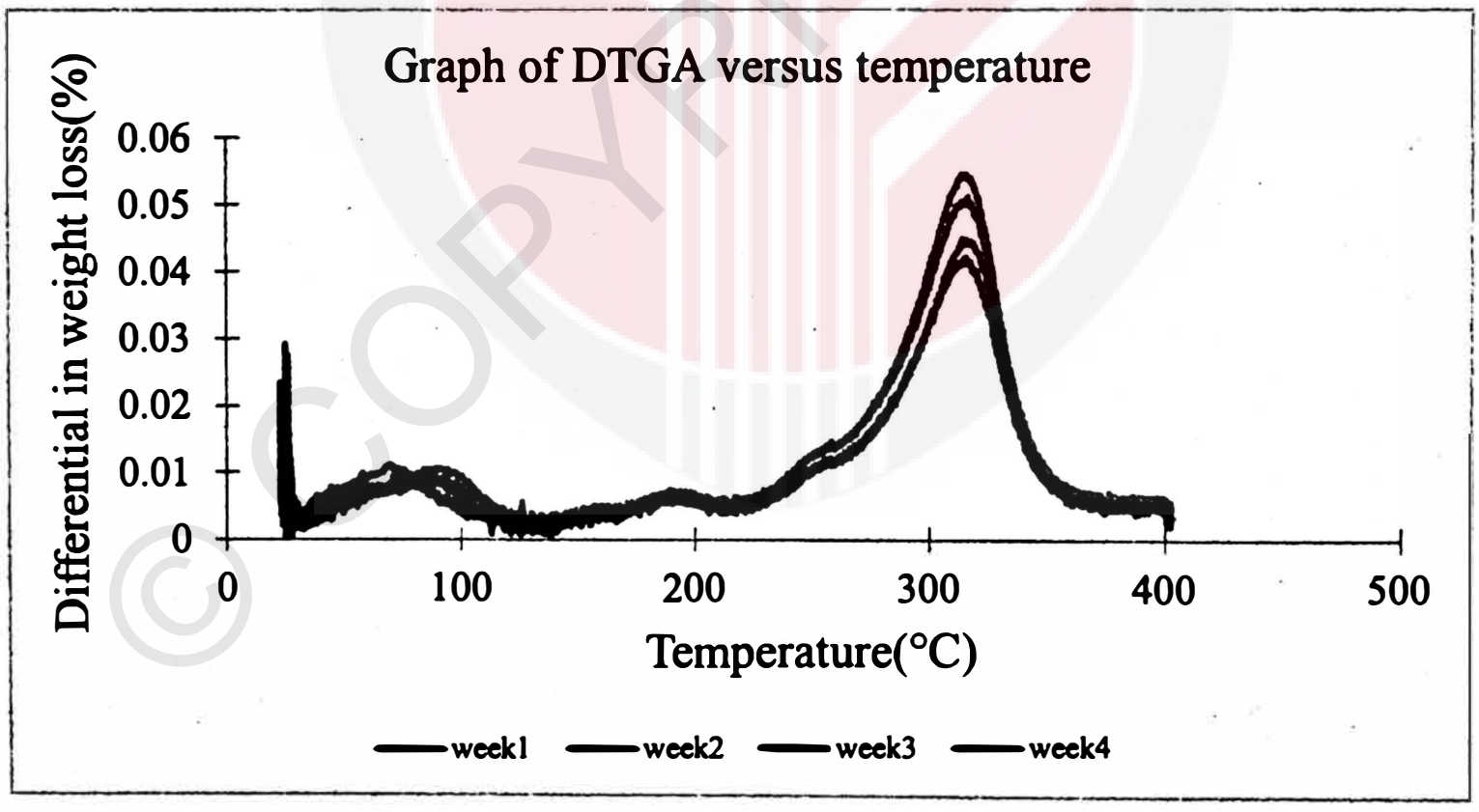


Figure 15 DTGA curve of encapsulated samples after storage of 28 days.

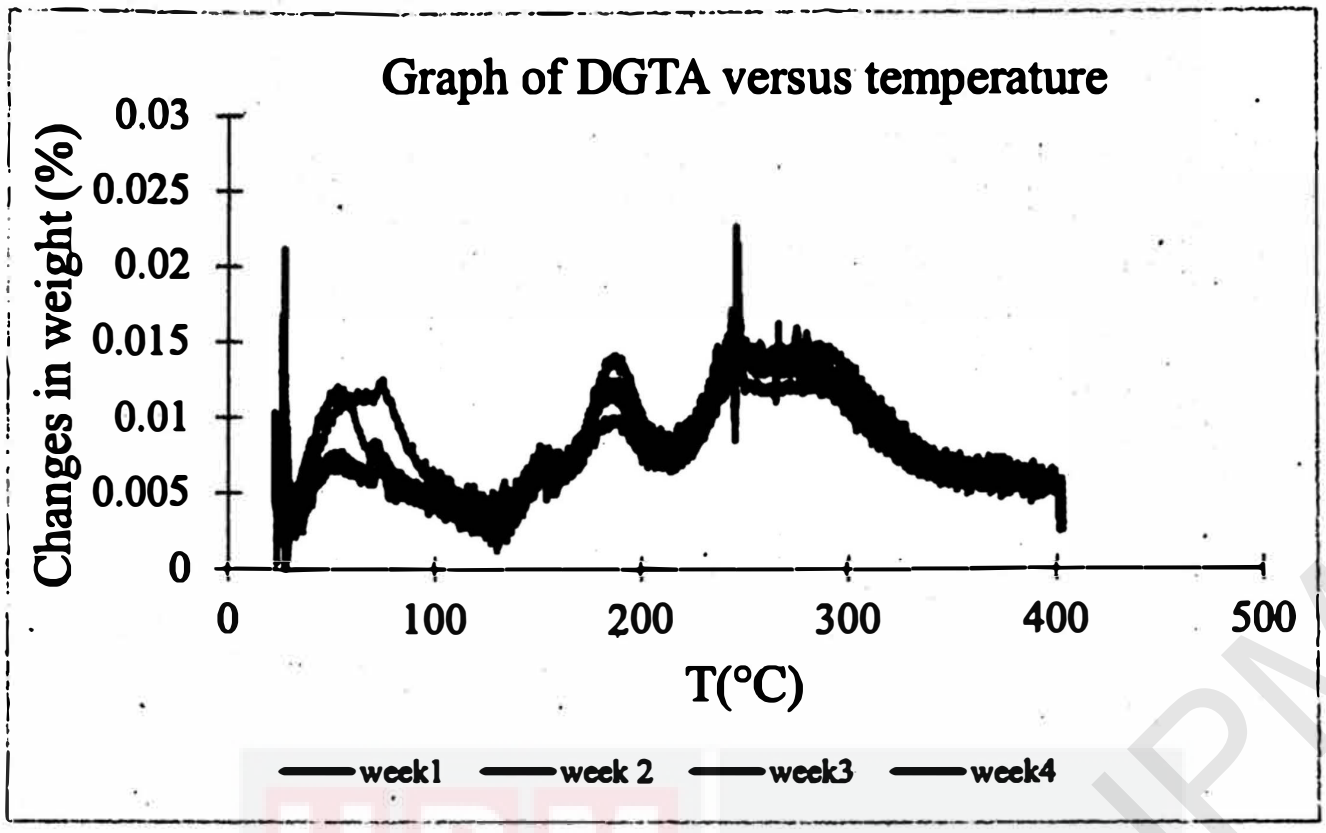


Figure 16 DTGA curve of non-encapsulated samples after storage of 28 days.

CHAPTER 5

CONCLUSION AND RECOMMENDATION

5.1 Conclusion

Jasmine flower possesses various bioactive compounds that can be extracted, electrospray and encapsulated using β-cyclodextrin for the application in the food industry, biomedical, active food packaging and so on. Electrospray drying is an important technology and simple because it can be applied in the atmospheric condition. There are two mechanisms involved in the electrospray drying which are solvent evaporation and Coulomb fission. In this study, process parameters of solvent concentration and relative humidity were manipulated in order to investigate the contribution of the parameters above towards the rate of drying. According to that equation, the drying rate is influenced by the parameters of the area of the initial droplet and relative humidity. In this case study, the rate of drying is affected by the concentration and humidity of surrounding air. The rate of drying is higher in 5wt% with humidity 0.0052 kg water vapour/ kg product when compare with higher concentration 25wt% with humidity 0.0061 kg water vapour/ kg product which is $1.12x10^{-13}$ kg/s and $1.02x10^{-13}$ kg/s respectively. This phenomena is most probably due to the crystalline bridges formed when the liquid evaporates. This was supported by applying the equation of evaporation rate where the similar trend is obtained based on

the graph plotted, the evaporation rate is increased when the time of evaporation is increased. The rate of evaporation for 25wt% is lower compared to the 5wt% due to the water proportion content in the solidified powder is decreasing when the process of evaporation taking place.

Next, the encapsulated and non-encapsulated jasmine compounds were evaluated with regard to morphology, encapsulation efficiency and thermal stabilities. The morphology of the encapsulated bioactive compound is spherical and agglomerate free which gives higher stability when compare to the non-encapsulated bioactive compound. Other than that, the efficiency of encapsulation was analysed by using FTIR and TGA. In the FTIR analysis, the encapsulated bioactive compound consisted similar peaks with cyclodextrin and non-encapsulated jasmine powder prove the occurrence of encapsulation. Besides, it also possess new bands in FTIR spectra prove that interaction formed between guest and host molecules. However, after storage of 28 days, the loss and degradation of bioactive compounds occurred even after encapsulated with cyclodextrin. While in the TGA analysis, the thermal degradation of encapsulated bioactive compounds shifted to the higher temperature when compared to the non-encapsulated. By comparing the loading capacity of the bioactive compounds in encapsulated and non-encapsulated samples, we can deduce that the bioactive compounds can be protected using CD as encapsulating agent via electrospray technique, however, due to the effect of storage, it will lead to the degradation of bioactive compounds by calculating the loading capacity in the encapsulated samples. When comparing with the thermal stability and shelf life of the encapsulated bioactive compound before and after storage of 28 days, the loading capacity of encapsulated

bioactive compounds was decreasing week by week but the difference is not very significant which is approximately 5.2%.

Therefore, cyclodextrin inclusion complex is successfully produced via electrospray technique with the aim of minimize the degradation and increase the stability of the bioactive compounds.

5.2 Recommendations

The electrospray drying technique can be carried out in ambient condition room with a control operating parameter such as temperature and humidity to prevent delocalization of electrostatic charge. The fluctuation of relative humidity and temperature may affect the formation of Taylor cone jet mode and cause the mechanism of Coulomb fission and solvent evaporation do not carry out efficiently. Besides that, the apparatus of the setup of electrospray should be avoided from electrical conductivity items to prevent the fluctuation of charge.

In this project, the concentration of bioactive compounds can be increased by changing the ratio of Jasmine flower extract and water, for example 5g of jasmine flower extract to 20ml of water to obtain 25wt% of jasmine suspension. During the preparation of encapsulated bioactive compounds, the duration of stir the cyclodextrin and the extracted solution can be increased to allow uniform mixing and encapsulation occur. Other than that, the solution also needs to cover with aluminium foil to prevent evaporation of the bioactive compounds.

The time of the electrospray also needs to prolong in order to get a higher deposition of jasmine powder on the aluminium foil for the analysis of FTIR and TGA. When the concentration of bioactive compounds increases, it is easier for the detection of peaks and the presence of bioactive compounds in the sample.

In this study, proton nuclear magnetic resonance (HNMR) can used to confirm the inclusion complexation is take place by determine the amount of bioactive compounds present in the encapsulated jasmine powder (Sambasevam, Mohamad, & Sarih, 2013). Moreover, this technique can be the supporting results for the TGA analysis to study the shelf life of the encapsulated bioactive compounds after 28 days of their storage by integrating the peak ratio of the characteristic chemical shifts corresponding to cyclodextrin and bioactive compounds.

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APPENDIX

Appendix 1 Field Emission Scanning Electron Microscopy (FE-SEM)

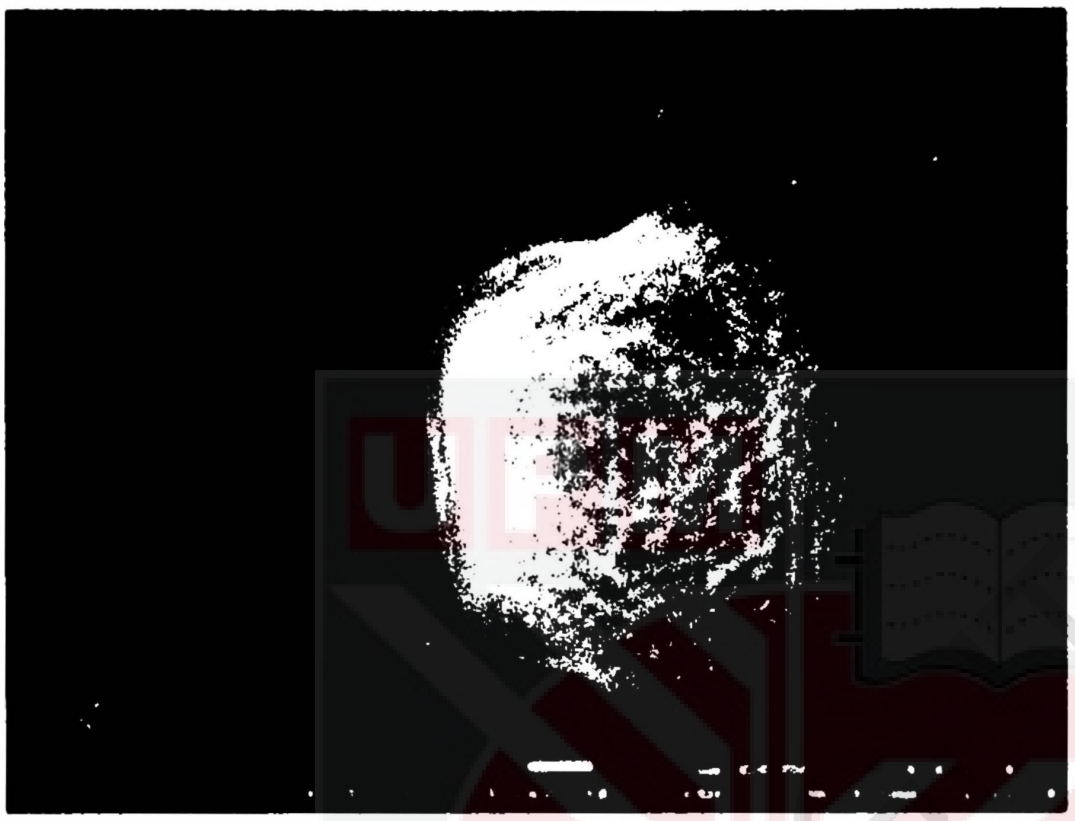

Figure A1: FE-SEM image of non-encapsulated

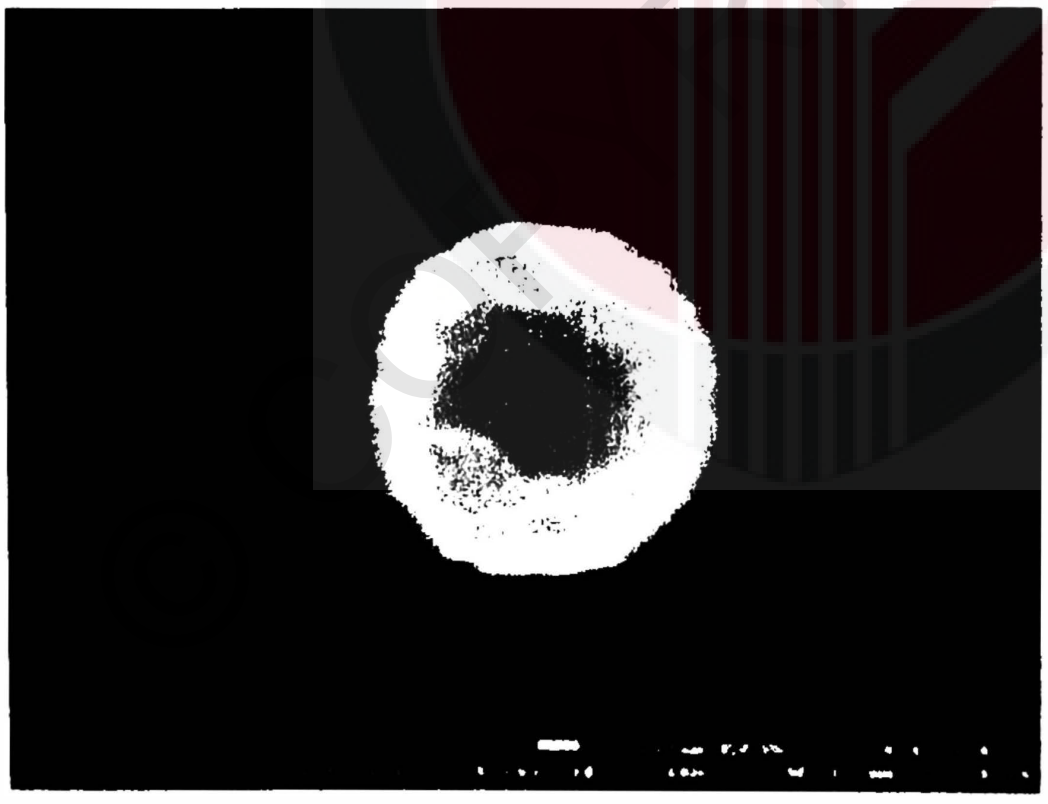

Figure A2: FE-SEM image of encapsulated jasmine powder

Appendix 2 Fourier Transform Infrared (FTIR)

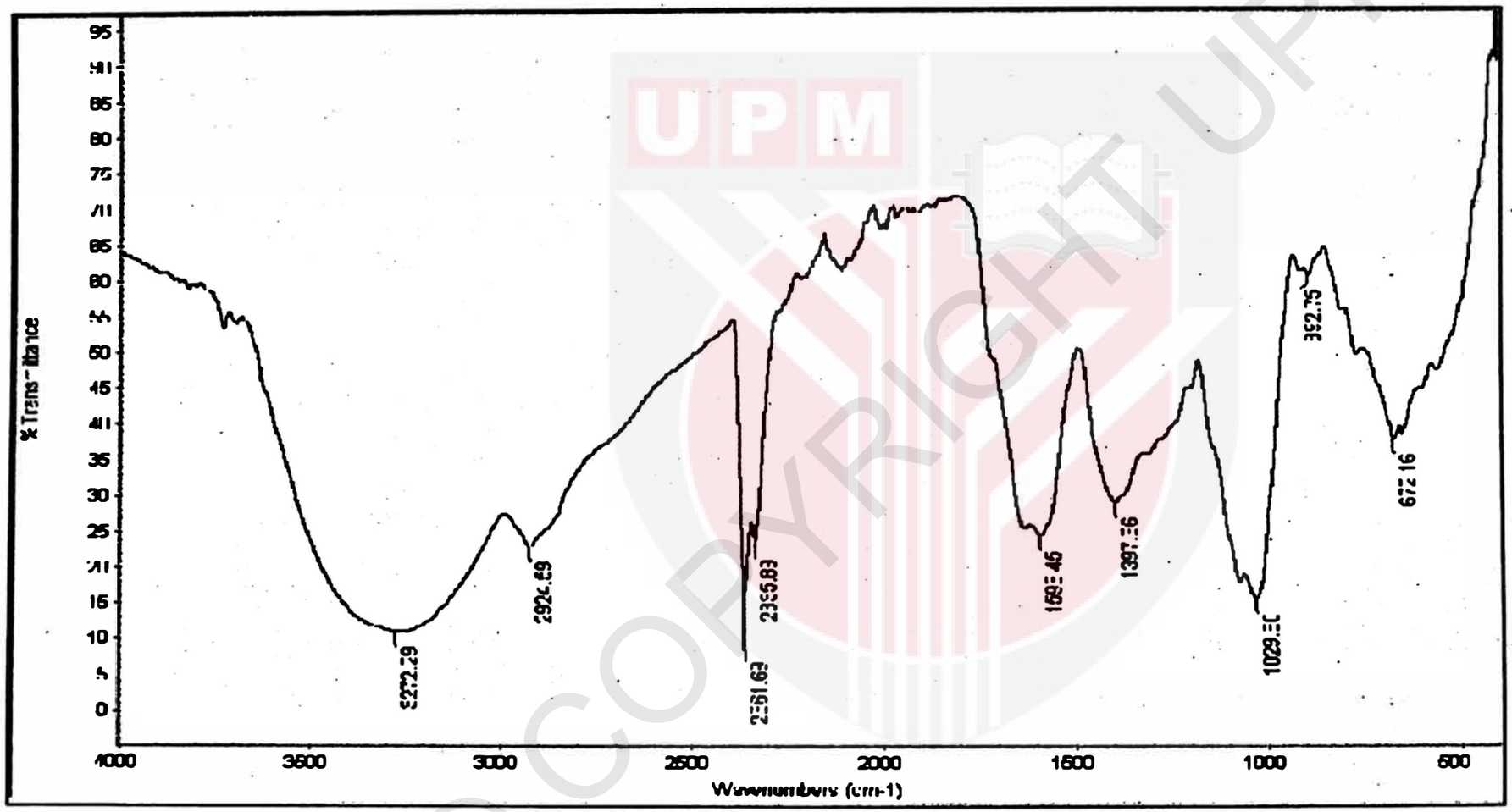

Figure B1: FTIR of non-encapsulated jasmine powder

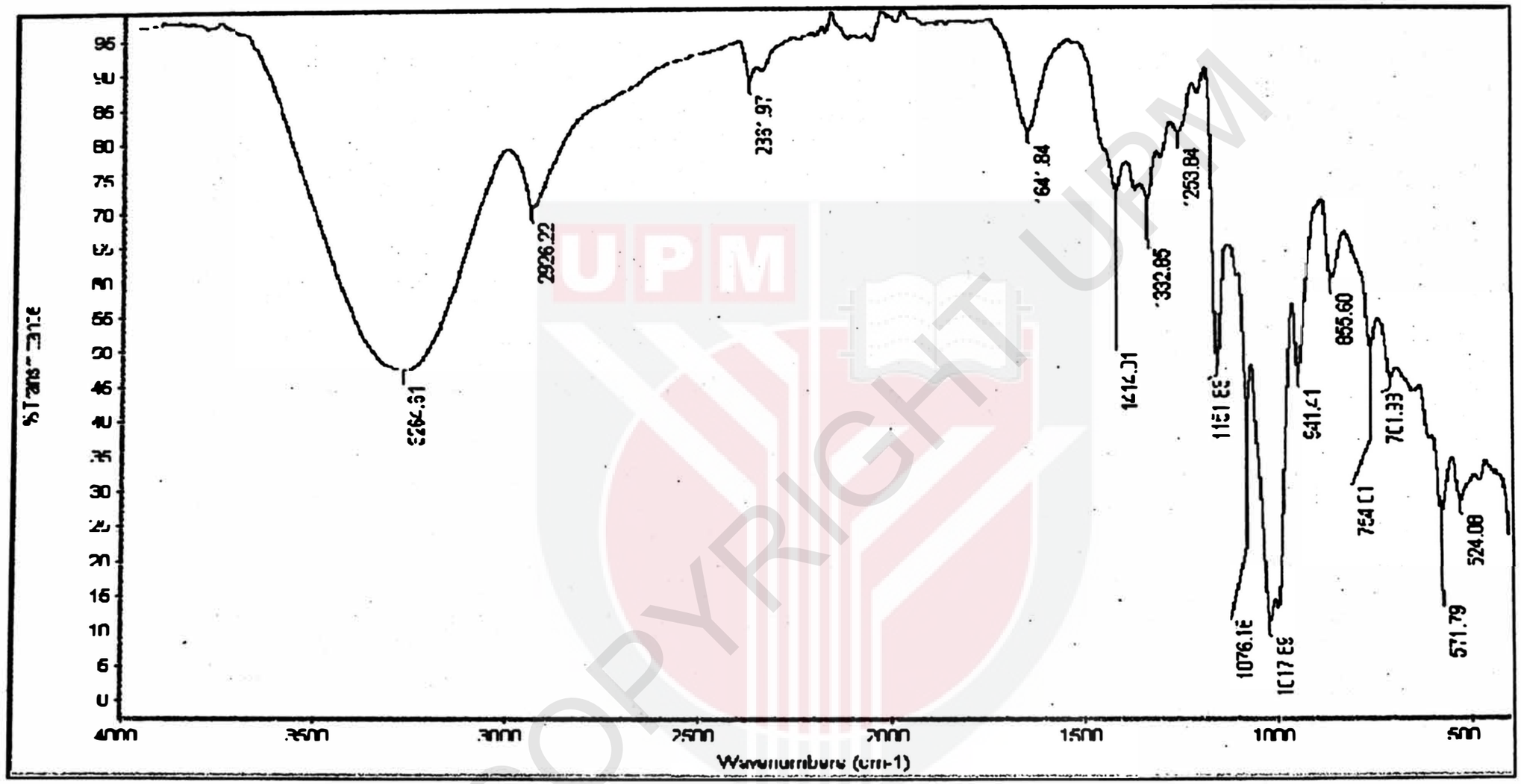

Figure B2: FTIR image of β-cyclodextrin

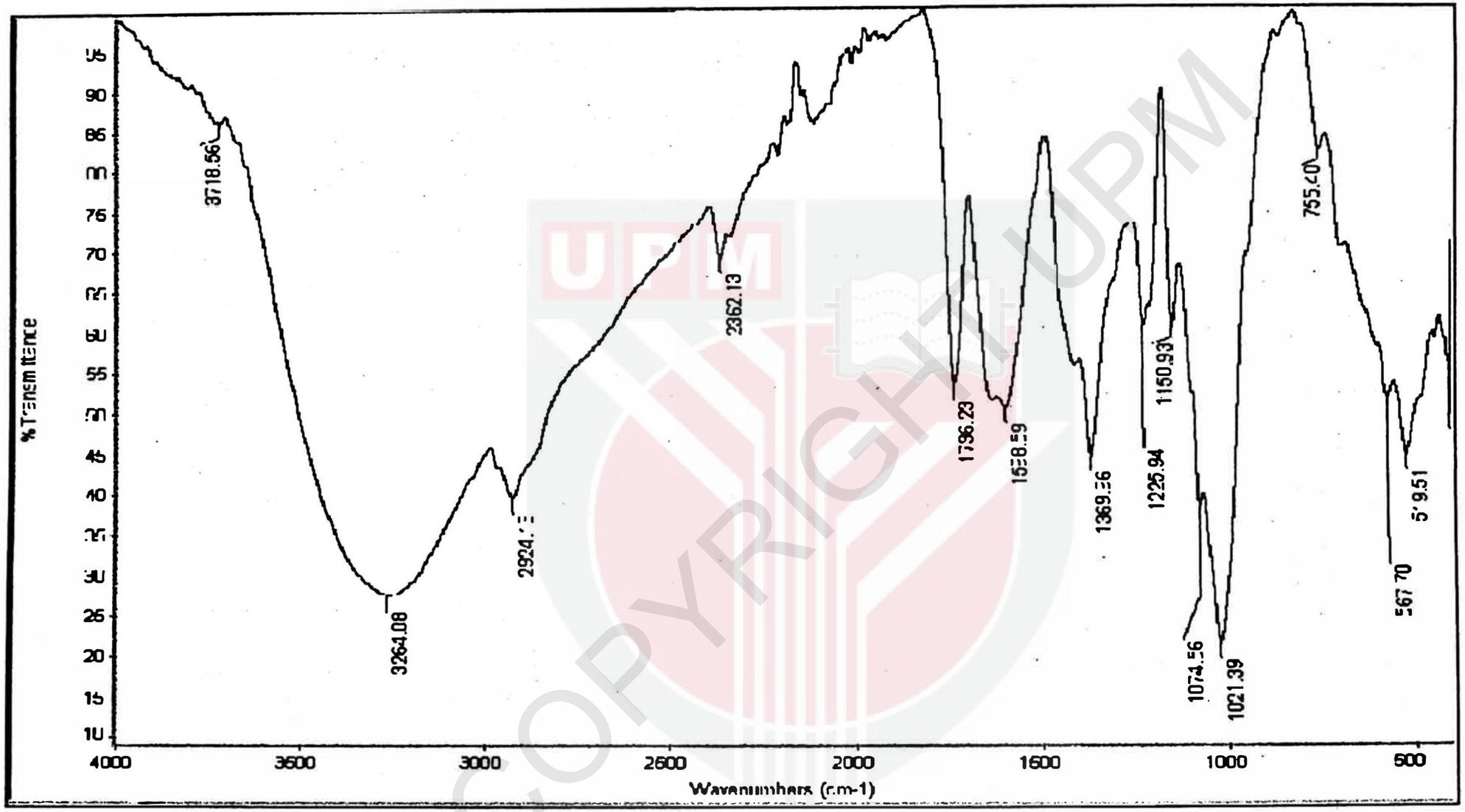

Figure B3: FTIR image of encapsulated jasmine powder before storage

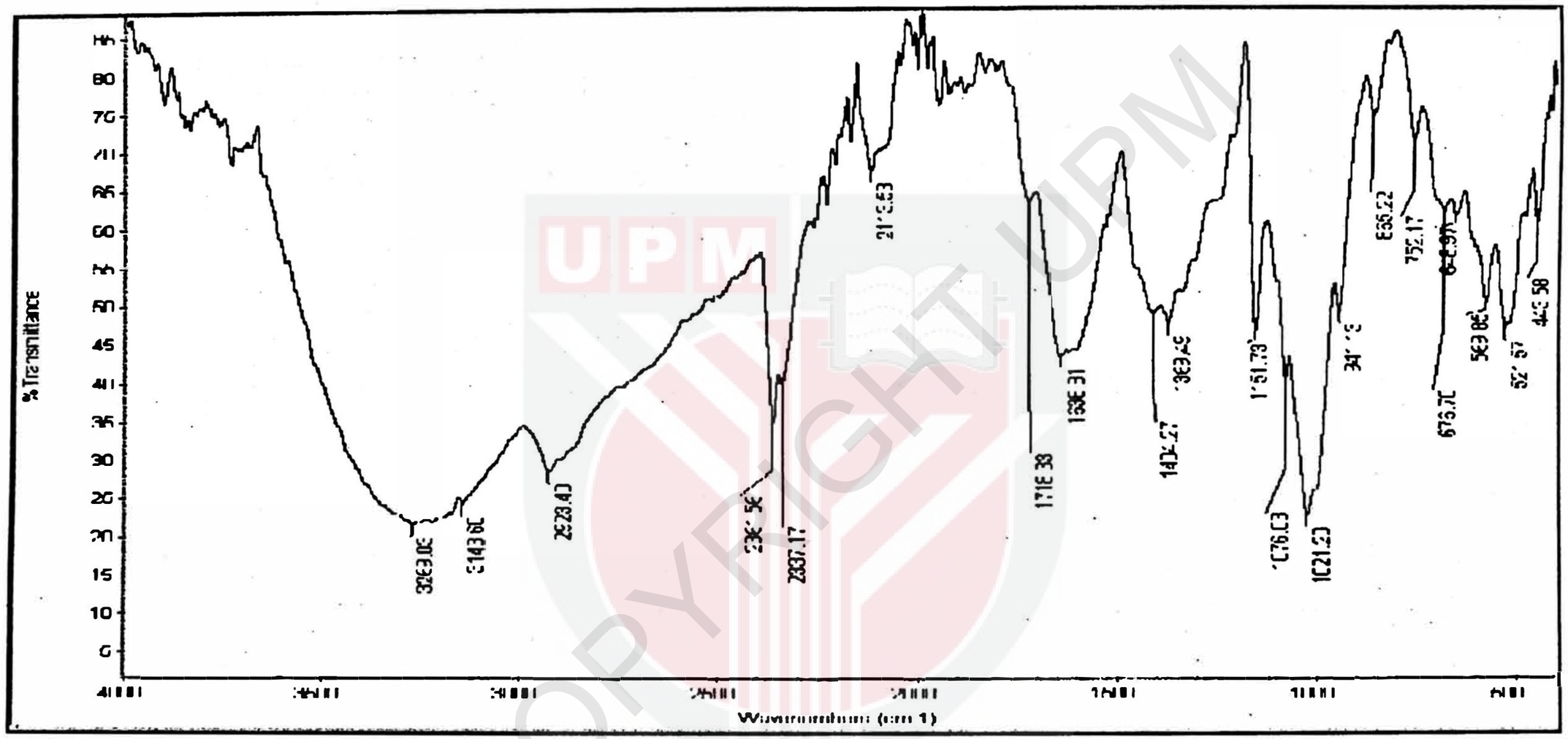

Figure B4: FTIR image of encapsulated jasmine powder week 2

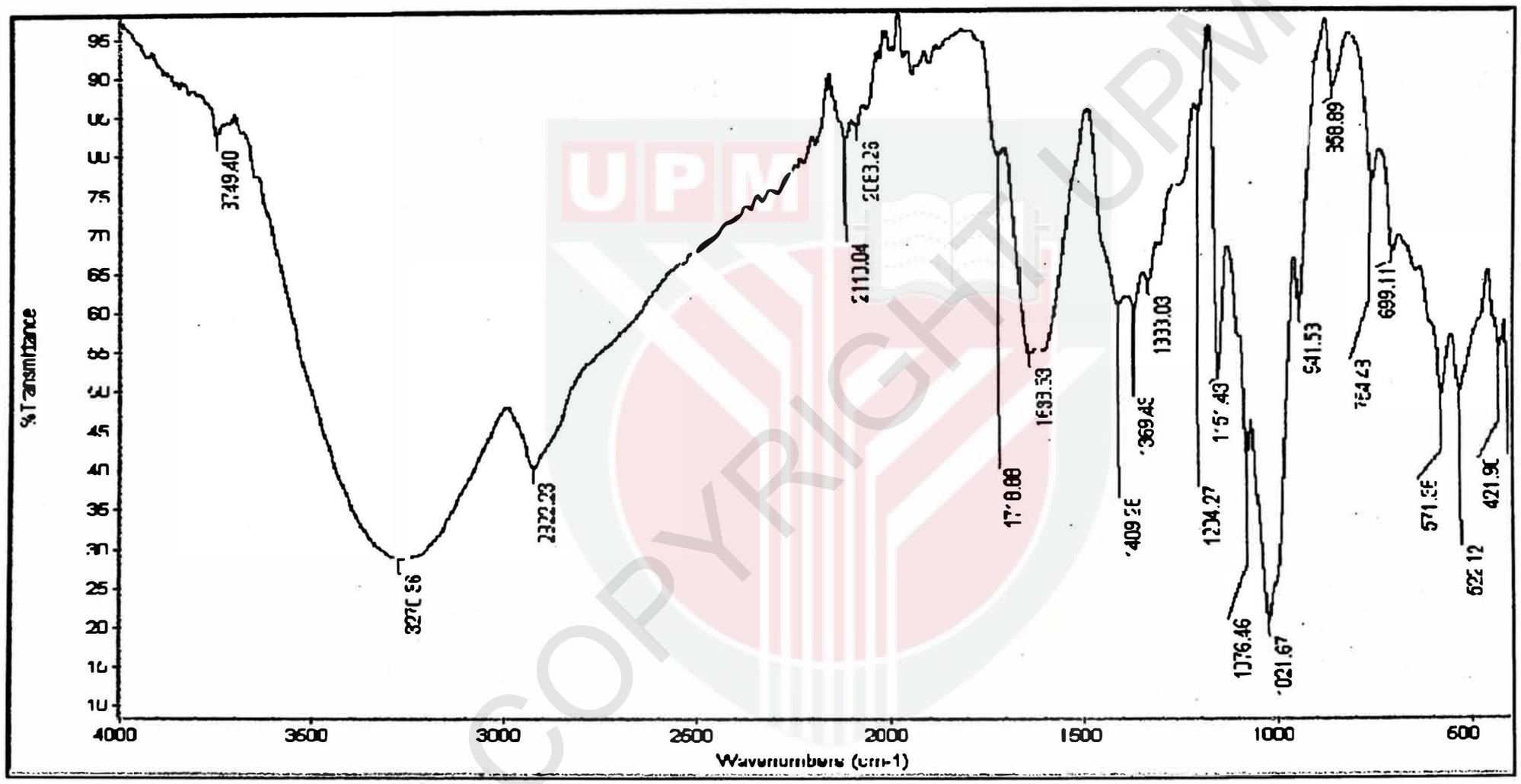

Figure B5: FTIR image of encapsulated jasmine powder week 3

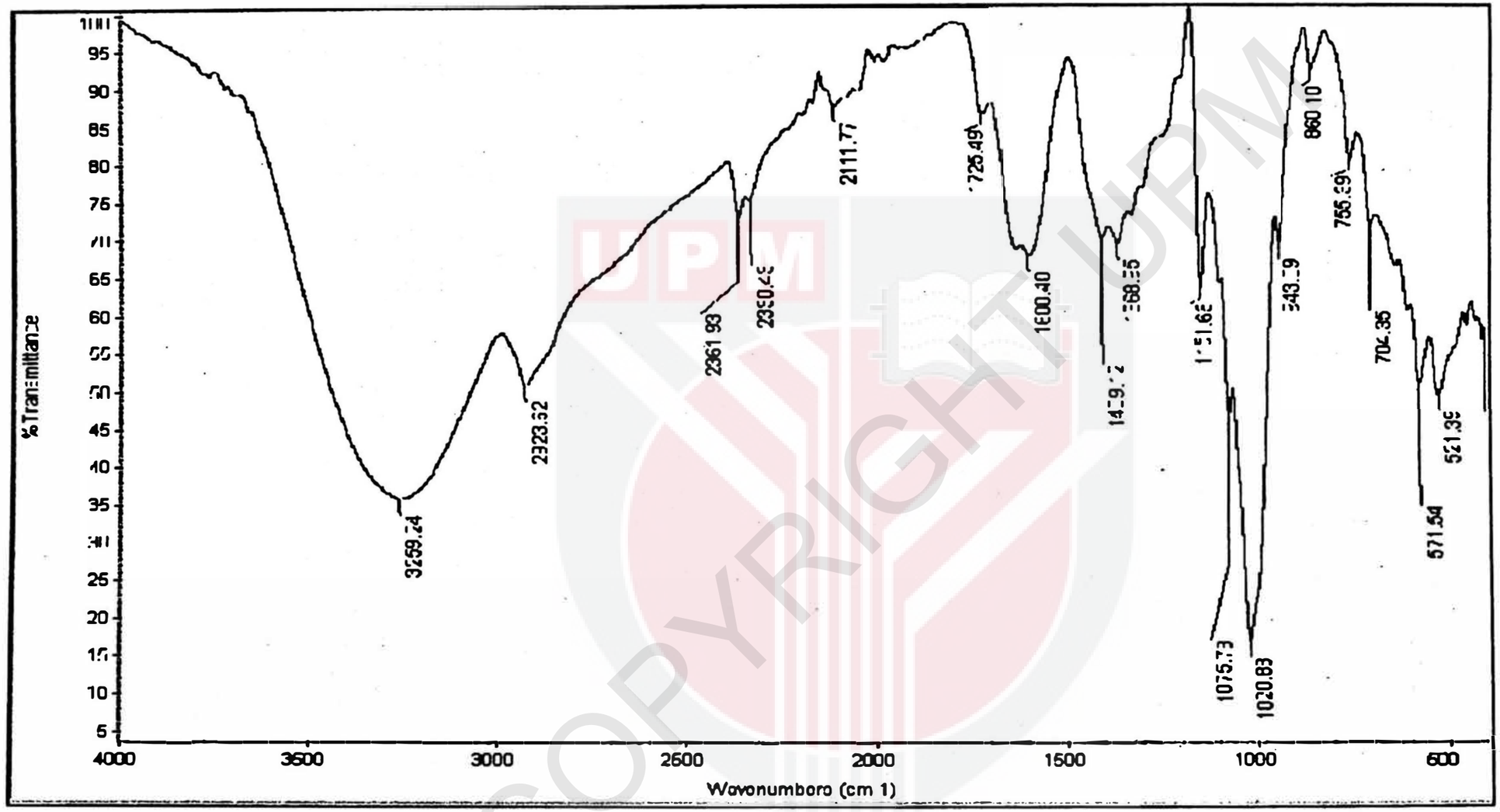

Figure B6: FTIR image of encapsulated jasmine powder after storage of 28 days

Appendix 3 Thermogravimetric Analysis (TGA)

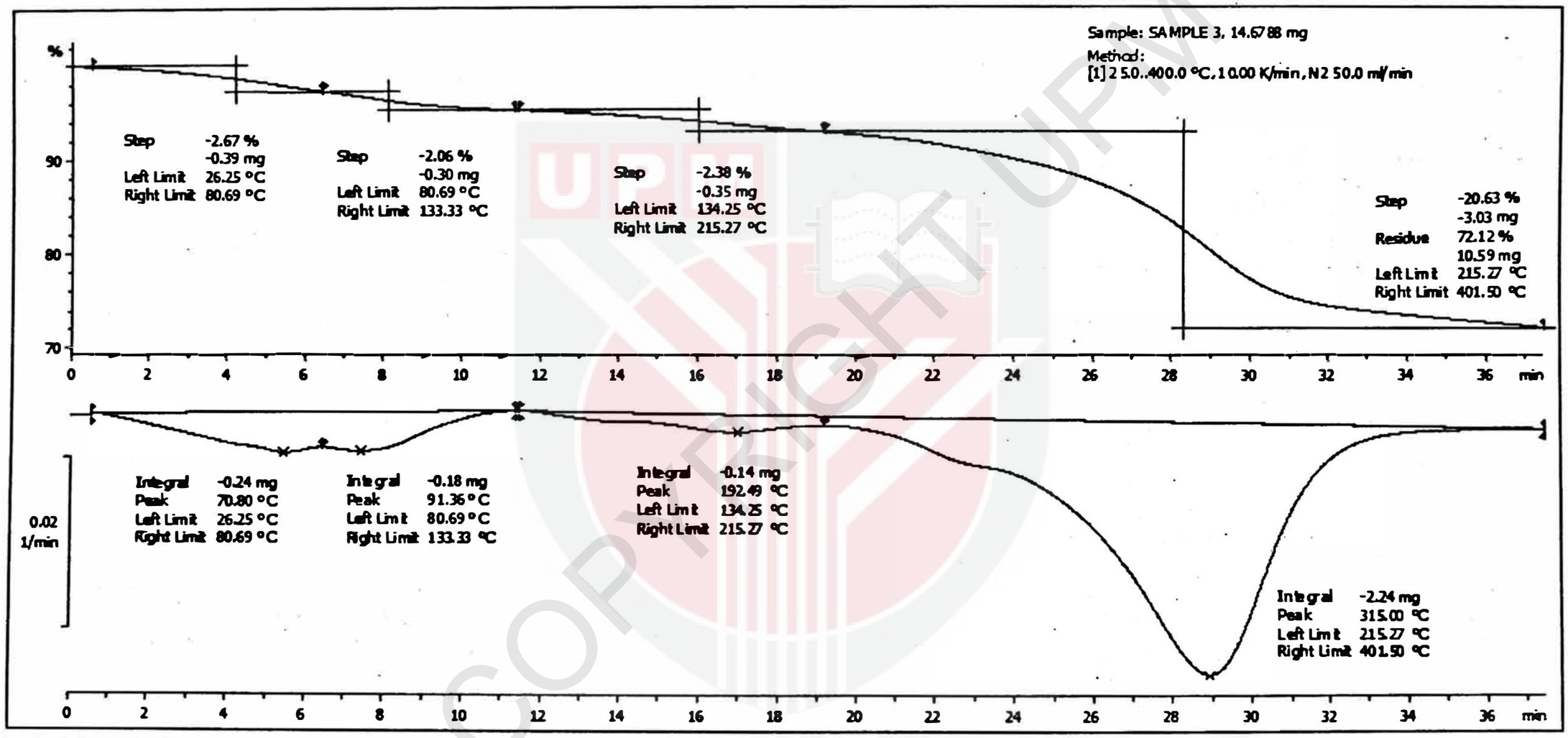

Figure C1: TGA curve of Week 1 encapsulated jasmine powder TGA curve

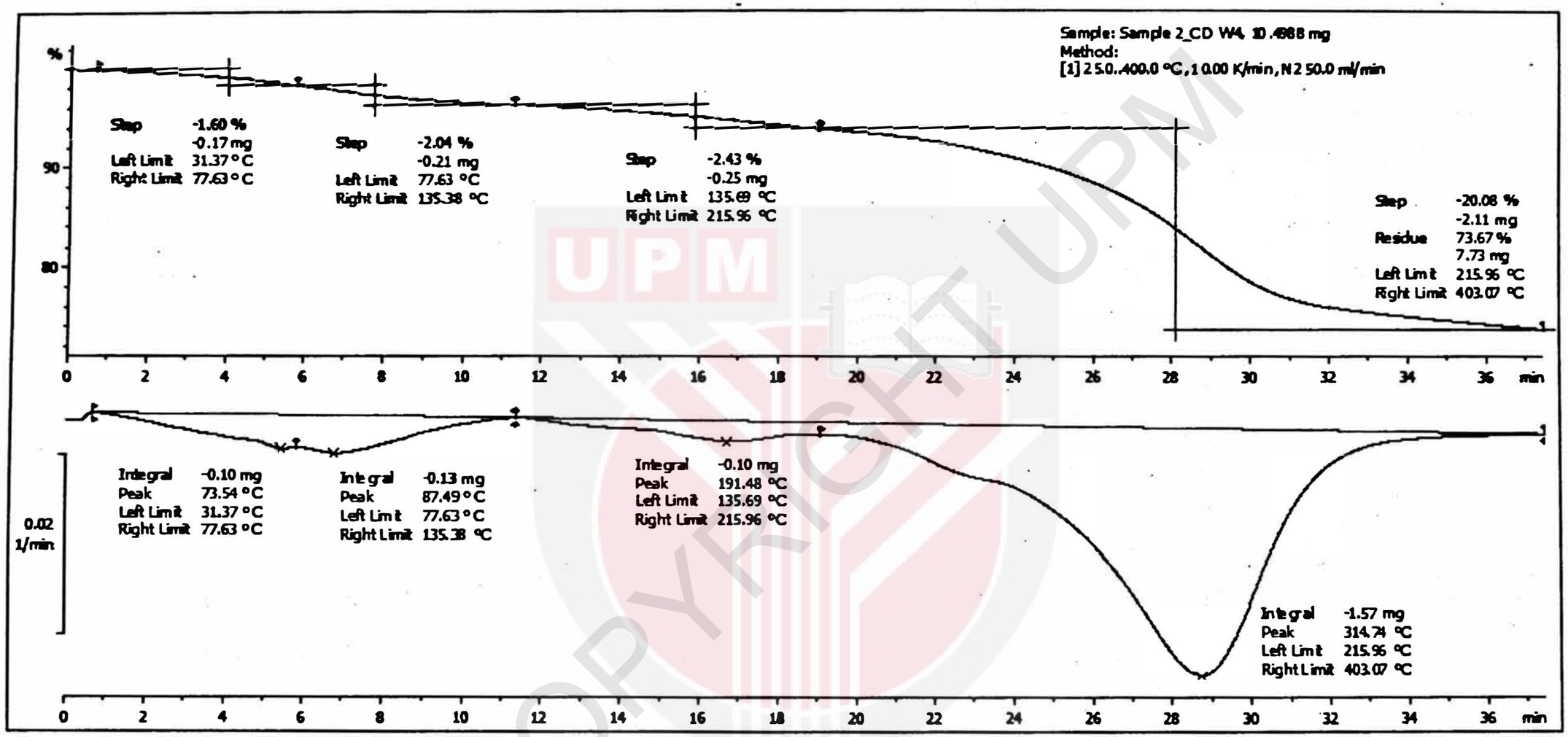

Figure C2: TGA curve of Week 2 encapsulated jasmine powder TGA curve

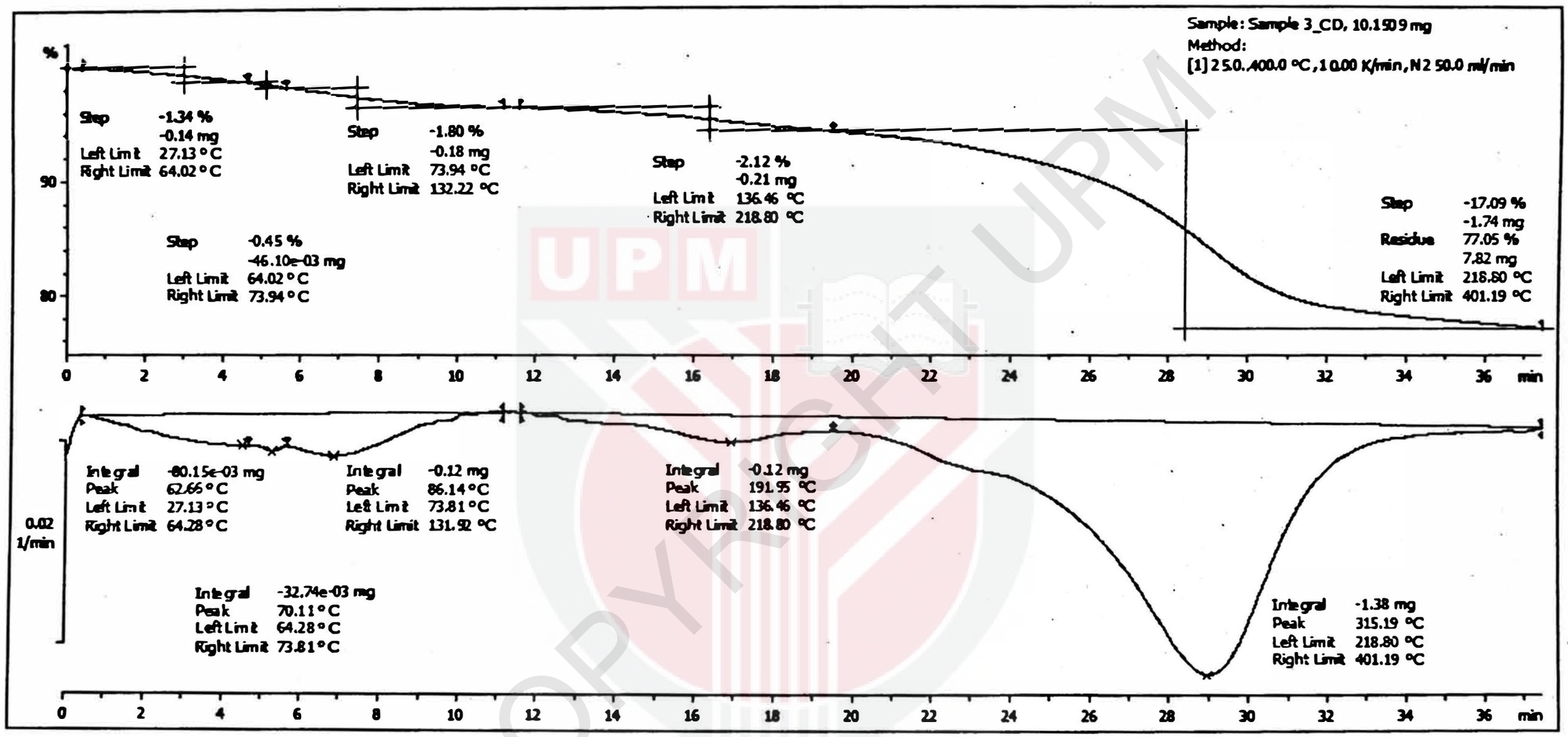

Figure C3: TGA curve of Week 3 encapsulated jasmine powder TGA curve

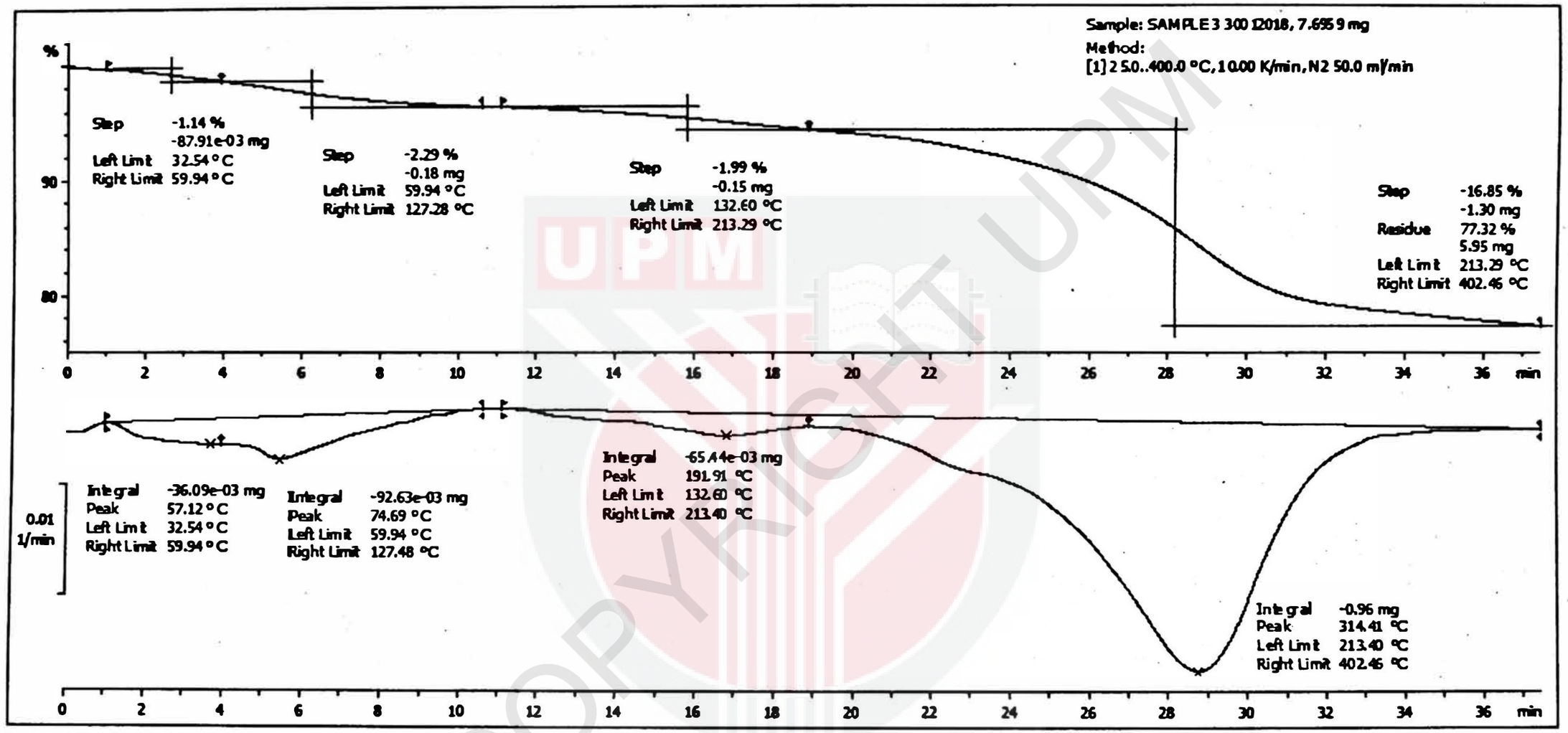

Figure C4: TGA curve of Week 4 encapsulated jasmine powder TGA curve

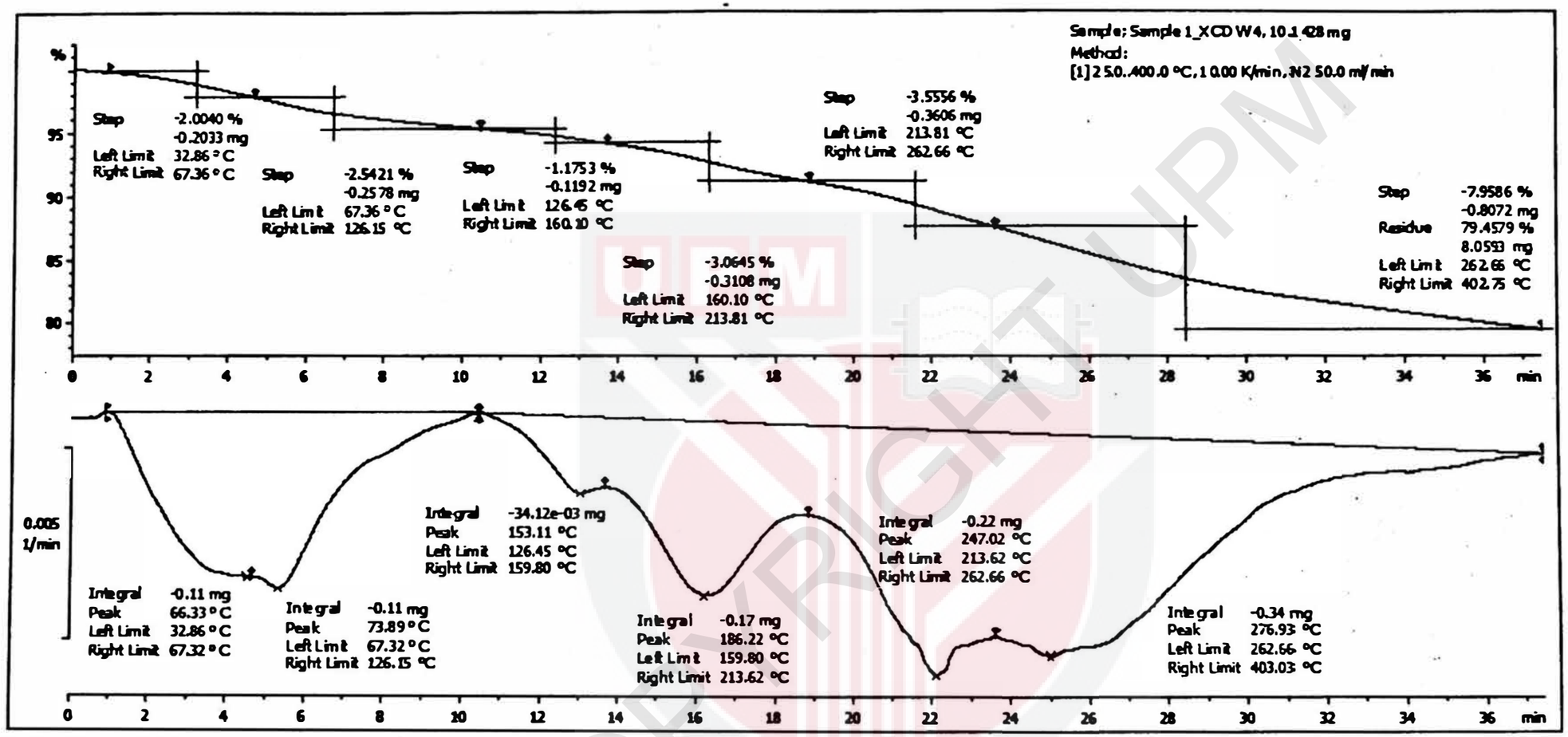

Figure C5: TGA curve of Week 1 non- encapsulated jasmine powder TGA curve

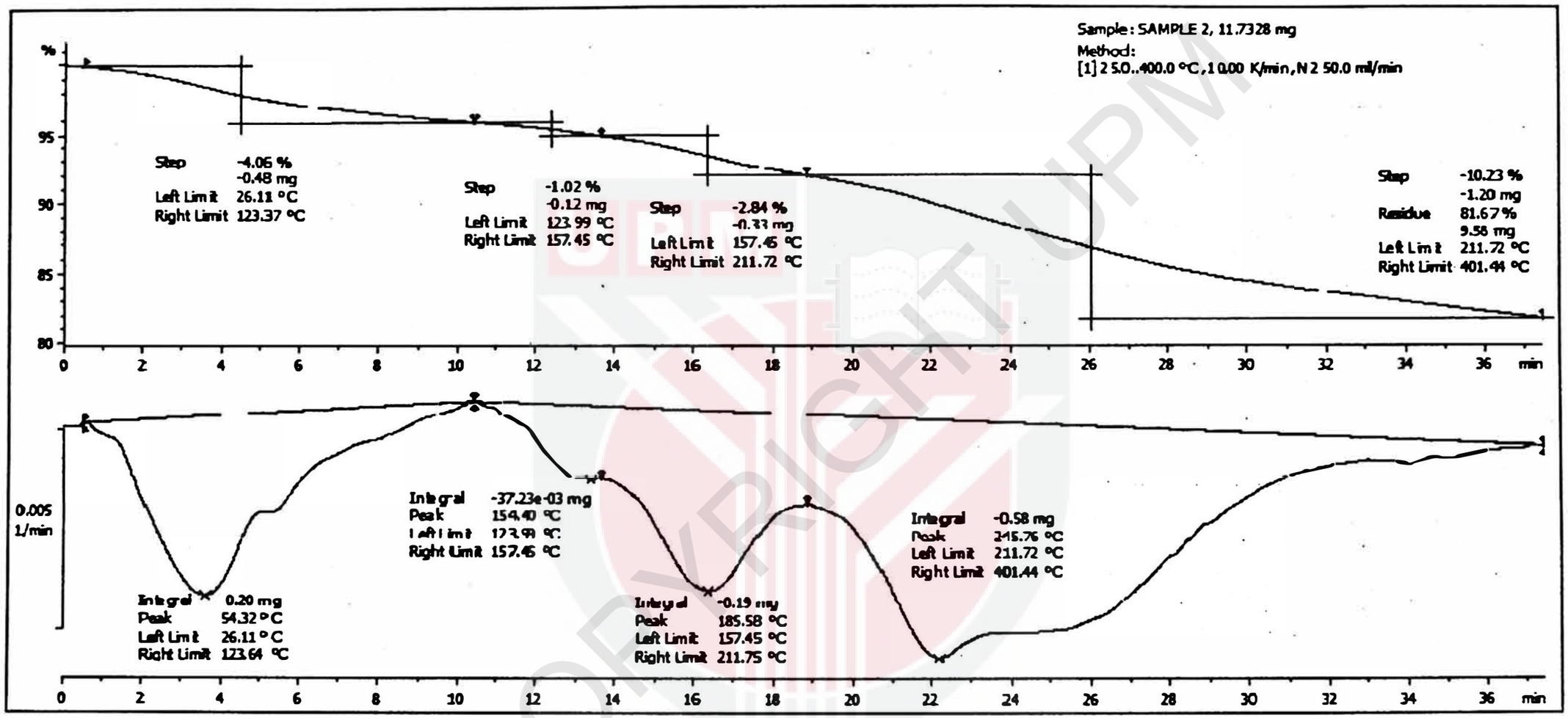

Figure C6: TGA curve of Week 2 non- encapsulated jasmine powder TGA curve

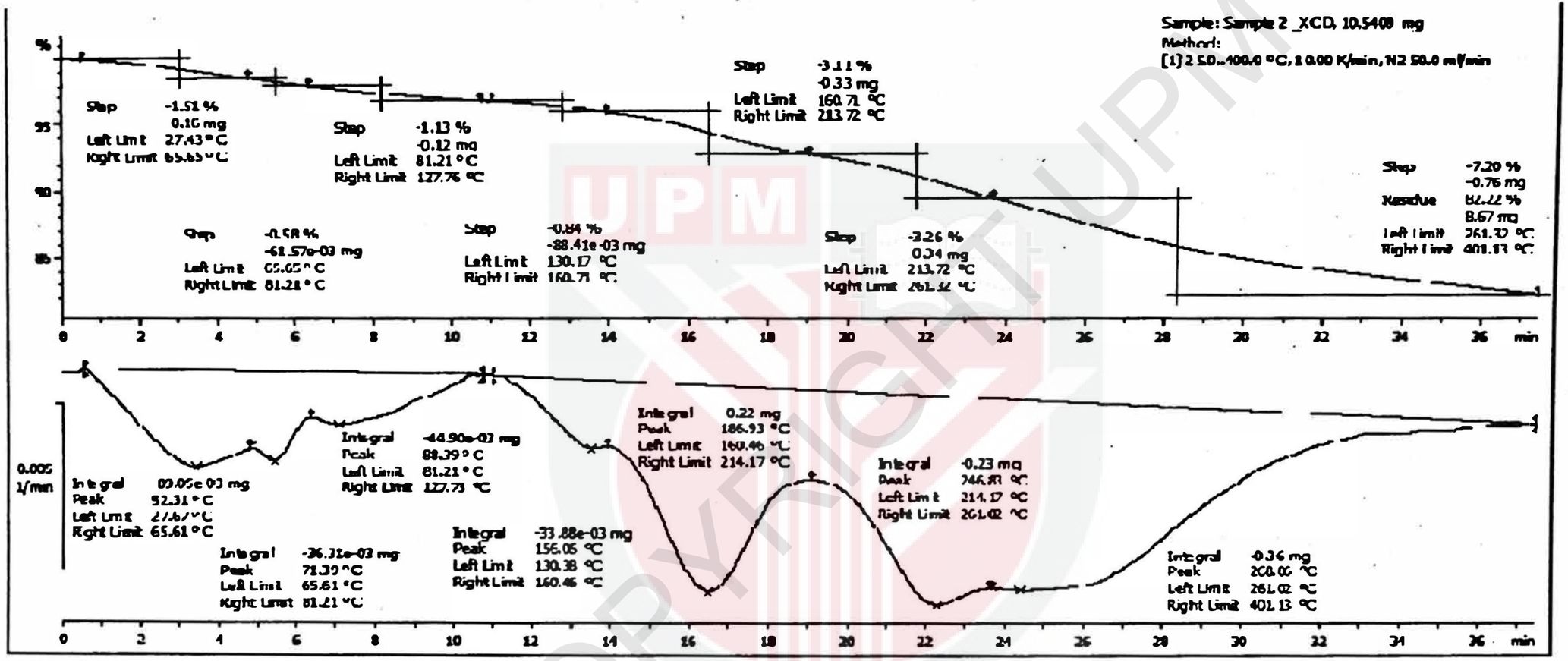

Figure C7: TGA curve of Week 3 non- encapsulated jasmine powder TGA curve

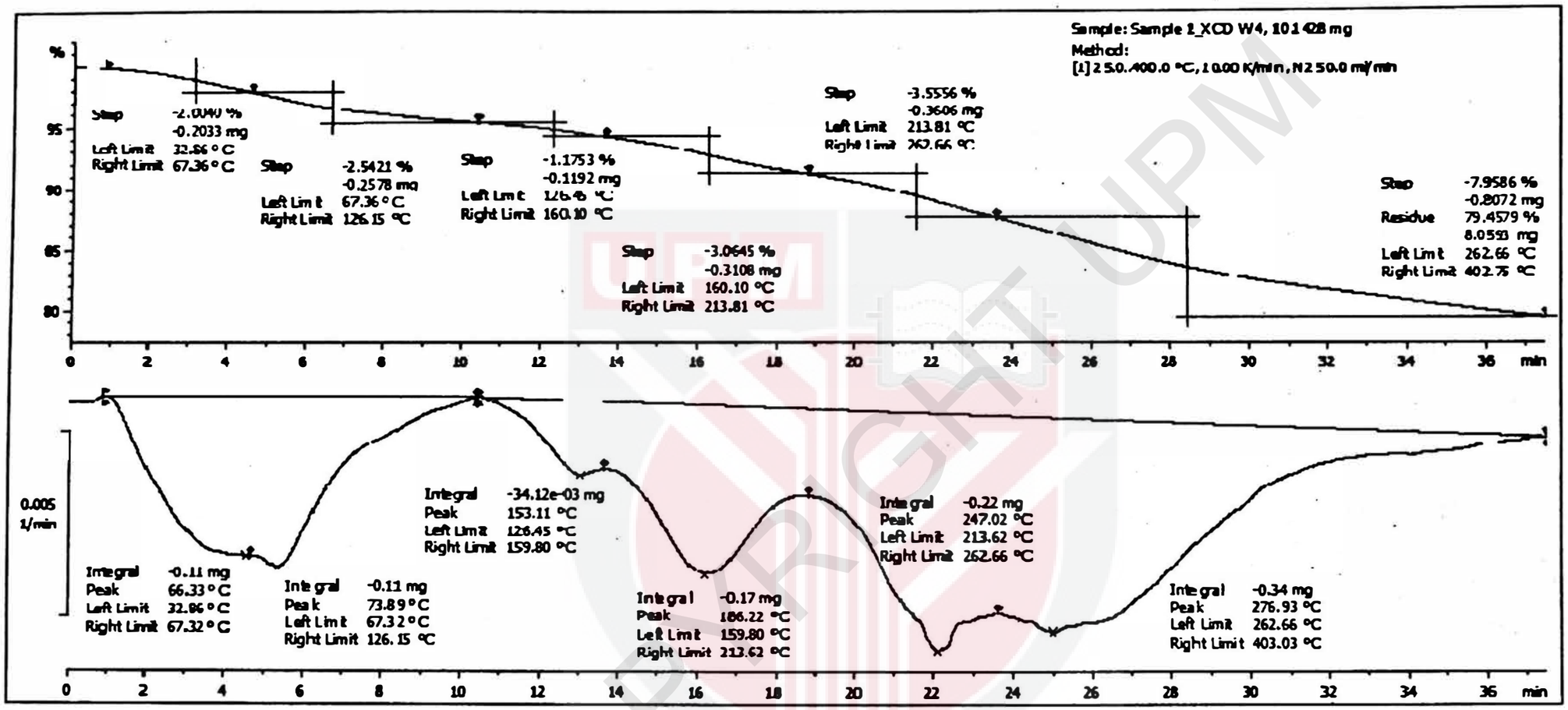

Figure C8: TGA curve of Week 4 non- encapsulated jasmine powder TGA curve