RESPONSE SURFACE OPTIMISATION OF PROCESS VARIABLES FOR ENCAPSULATION OF CUMIN OIL BY SPRAY DRYING

By

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(2016-18-003)

THESIS

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DECLARATION

I hereby declare that this thesis entitled **"Response surface optimisation of process variables for encapsulation of cumin oil by spray drying"** is a bonafide record of research work done by me during the course of research and the thesis has not previously formed the basis for the award to me of any degree, diploma, associateship, fellowship or other similar title, of any other University or Society.

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SYMBOLS AND ABBREVATIONS

-	:	Minus
%	:	Per cent
+	:	Plus
<	:	Less than
~	:	In the range of
>	:	Greater than
°C	:	Degree centigrade
&	:	And
/	:	Per
=	:	Equal to
µm	:	Micro metre
a*	:	Greenness or redness
ANOVA	:	Analysis of variance
AOAC	:	Association of Official Analytical Chemists
a_w	:	Water activity
b*	:	Blueness or yellowness
cm	:	Centimeter
CTCRI	:	Central Tuber Crops Research Institute
DC	:	Direct current
DE	:	Dextrose equivalent
EE	:	Encapsulation efficiency

et al	:	And others
etc.	:	Etcetera
Fig.	:	Figure
g	:	Gram
GA	:	Gum arabic
g/cm^3	:	Gram per centimetre cube
g/min	:	Gram per minute
GC	:	Gas chromatography
GC-MS	:	Gas chromatography mass spectroscopy
h	:	Hour (s)
ha	:	Hectare
HP	:	Horse power
I.D	:	Inner diametre
i.e.	:	That is
KAU	:	Kerala Agricultural University
KCAET	:	Kelappaji College of Agricultural Engineering and Technology
Kg	:	kilogram
Kg/h	:	Kilogram per hour
Kw	:	Kilo Watt
L*	:	Lightness or darkness
Ltd.	:	Limited
LCD	:	Liquid crystal display

m	:	Metre
m^2	:	Metre square
m.c.	:	Moisture content
MD	:	Maltodextrin
Min	:	Minute (s)
ml	:	Milli litre
ml/h	:	Milliliter per hour
ml/min	:	Milliliter per minute
mm	:	Millimetre
nm	:	Nanometre
No.	:	Number
O.D	:	Outer diameter
Pa	:	Pascal
rpm	:	Revolution per minute
Rs.	:	Rupees
RSM	:	Response Surface Methodology
s	:	Second (s)
SEM	:	Scanning Electron Microscope
Sl.	:	Serial
SS	:	Stainless steel
USA	:	United States of America
viz	:	Namely

w.b.	:	Wet basis
Wt.	:	Weight
w/v	:	Weight by volume
w/w	:	Weight by weight

Introduction

CHAPTER I

INTRODUCTION

Spices are the main flavouring agents in food and were in use since the birth of civilisation. It is mainly used for providing aroma, taste, flavour, texture and colour to food, and can also be used as preservative and can provide nutritional, and health benefits. Spices are essential in culinary and medicinal purposes and hence occupy a place in all the cultures of the world. Spices can be from root (ginger), bark (cinnamon), buds (clove), berries (grains of pepper), leaf (e.g. bay leaf), seeds (cumin), or even the stigma of flower (saffron) (Viuda-Martos *et al.*, 2011).

Cumin (*Cuminum cyminum Linn.*) is one of the important commercial seed spices which belong to the *Umbellifereae* family. Cumin is mainly valued for its aroma, medicinal and therapeutic properties. It is a characteristic spice of the oriental cuisine and a major ingredient of curry powder (Acimovic *et al.*, 2016). These spices have considerable demand as a spice all around the globe, particularly in areas where people prefer spicy foods. The plant is native to the Mediterranean region and North Africa. It is a very popular spice in America, Burma, India and Indonesia. In India, the area under cumin cultivation in the year 2015-2016 was 808230 ha and the production was 503260 tonnes (Spices Board, 2015). The main cumin cultivating states in India are Rajasthan, Gujarat and Madhya Pradesh (Bankar, 2011).

Indian cumin finds worldwide use in food, liquor, medicine, beverages, perfumery and toiletries. After pepper, cumin is considered to be the most important spice in the world (Sowbhagya, 2013). Various studies have shown that *Cuminum cyminum* can impart antimicrobial, anti-inflammatory, anticancer, insecticidal, antioxidant, analgesic, antiplatelet aggregation, antidiabetic, hypotensive, contraceptive, bronchodilatory, anti-amyloidogenic, immunological, aldose reductase , anti-osteoporotic, alpha-

glucosidase and tyrosinase inhibitory effects, protective and central nervous effects (Al-Snafi, 2016).

The most important chemical component of cumin seed is essential oil content. This essential oil content ranges from 2.5 to 4.5% and is pale to colourless depending on age and regional variations (Nadeem and Riaz, 2012). The cumin oil obtained by steam distillation is mainly used to flavour alcoholic beverages, desserts and condiments. In addition, it can also be used as a fragrant component of perfumes, lotions and creams (Singh *et al.*, 2017).

The biological activity of the oil is lost due to the volatilisation or degradation of active compounds owing to direct exposure to heat, humidity, light, or oxygen. Hence these oils have to be protected from all these factors. Encapsulation is the most suitable method which will protect the essential oil and flavour ingredients from the liquid form to solid form by coating agents. Thus the products become very easy to handle. In addition, encapsulation prevents lumping, improves flowability, compression and mixing properties, reduces core particle dustiness and modifies particle density (Ravichandran *et al.*, 2014). This technique will entrap sensitive ingredients, such as volatile and labile flavours, into solid carriers and thereby increases their protection, reduces evaporation and controls their release during storage and application (Hussein *et al.*, 2016). Encapsulated flavours enable a high sensory impact of the final food product.

Microencapsulation and nanoencapsulation are the two encapsulating techniques commonly used. If the encapsulated product have sizes in micrometre or larger, then it is called microencapsulation. Most of the microencapsulated products have diameter between 1 to 1000 μm (Suganya and Anuradha, 2017). Nanoencapsulation is the application of encapsulation in the nanometre scale with films, layers, coverings, or simply

microdispersion. Nanoparticles are colloidal-sized particles with diameters ranging from 10 to 1,000 nm (Ezhilarasi *et al.*, 2013). Compared to micron-sized carriers, nanocarriers can provide larger surface area and have the potential to increase bioavailability, enhance solubility, improve controlled release, and enable precision targeting of the encapsulated materials to a greater extent (Mozafari *et al.*, 2008).

The selection of appropriate encapsulating material is very important as it influences the encapsulation efficiency and microcapsule stability (Estevinho *et al.*, 2013). The right combination of wall materials can produce preferred characteristics in the encapsulated materials. A wide variety of natural and synthetic polymers can be used as the wall materials in the encapsulation process. Usually natural food-grade compounds are opted for minimising the potential toxic effects associated with ingestion. Some of the natural polymers usually used are, gums (e.g., carrageenans, guar gum and gum arabic), carbohydrates (e.g., cellulose derivatives, chitosan, cyclodextrins, dextrose, lactose, maltodextrins, sodium alginate and starches) and proteins (e.g., albumin, casein, collagen, egg, gelatine, soy and whey) (Arpagaus *et al.*, 2017). Among these coating materials, gum arabic (gum acacia) has been the standard of excellence as a flavour encapsulating material for many years. It is an excellent emulsifier, bland in flavour and provides good retention of the volatile compounds during the drying process (Kausadikar *et al.*, 2015). But increased cost, limited supply and variations in the quality have restricted the use of gum arabic for encapsulation purpose (Kanakdande *et al.*, 2007). Maltodextrin is a possible option to use as encapsulation agent as it is a good matrix former and can provide protection against oxidation, permit increased solid content with low viscosity and is economical, besides other advantages (Prince *et al.*, 2014). Starch is widely distributed renewable biopolymer in nature having a huge diversity of industrial applications. It is also one of the widely used wall material in encapsulation process (Hoyos-Leyva *et al.*, 2016).

Various encapsulation techniques used include spray chilling or spray cooling, spray drying, fluidised bed coating, extrusion coating, coacervation, liposome entrapment, inclusion complexation, rotational suspension separation and centrifugal extrusion (Gibbs *et al.*, 1999). Among all these techniques, spray drying is most extensively used in the food industry.

Spray drying is a rapid, continuous, scalable, reproducible and cost-effective process employed in the production of dry powders from a fluid material. In a spray dryer the liquid stream is sprayed through a nozzle into a hot vapour stream and it will separate the solute or suspension as a solid and the solvent into a vapour. The solid thus formed is usually collected in a cyclone or a drum. The nozzle makes the liquid droplets as small as possible for maximising the heat transfer and rate of water vaporisation. The spray drying technique produces good quality final product with reduced water activity and weight resulting in easy storage and transportation (Phisut, 2012). Most often the spray drying technique is considered as a dehydration process, but it can also be applied for the encapsulation of active materials when it entraps the core compound within a protective matrix, which is essentially nonreactive to the core materiel which is being, encapsulated (Re, 1998).

From the reviews it was evident that the protocols for scientific study on microencapsulation and nanoencapsulation of cumin oil by spray drying was lacking in the literatures. Hence this study was considered quite appropriate that intended to optimise the process protocols for the production of encapsulated cumin oil.

In light of the above points, the present investigation entitled "Response surface optimisation of process variables for encapsulation of cumin oil by spray drying" was undertaken at Kelappaji College of Agricultural Engineering and Technology (KCAET), Tavanur, Kerala with the following objectives.

1. To optimise process parameters for microencapsulated cumin oil
2. To optimise process parameters for nanoencapsulated cumin oil
3. To compare the optimally produced microencapsulated and nanoencapsulated cumin oil

Review of Literature

CHAPTER II

REVIEW OF LITERATURE

This chapter deals with the research findings reported by other researchers related to microencapsulation and nanoencapsulations of similar commodities and its quality evaluation.

2.1 Cumin

Cumin (*Cuminum cyminum L.*) is an annual plant in the *Umbelliferae* family. The cumin seeds are an essential ingredient of curry and chili powder, and commercially used for preparing pickles, sausages, meats, chutney and cheeses (Tassan and Russell, 1975).

India contributes about 90% of the world cumin production. Gujarat and Rajasthan are the major states which cultivate cumin in India (Shivakumar *et al.*, 2010).

Cumin seeds possess warm, heavy, spicy, and curry like flavour, which is dominated by the presence of flavouring constituent cuminaldehyde. The presence of volatile oil in the cumin seeds imparts a peppery astringent flavour with slight citrus overtones. Composition of cumin varies with the growing region and the climatic conditions (Sowbhagya, 2013).

Cumin is a commonly used condiment mainly to add characteristic aroma and flavour to food. Various bioactive compounds are present in the cumin seeds belonging to different categories of cumin aldehydes, polyphenols, terpenoids and flavonoids (Sultan *et al.*, 2014).

Cumin seeds play a major role as a characteristic spice in the oriental cuisine and contribute to one of the major ingredients of curry powder (Acimovic *et al.*, 2016).

According to Al-Snafi (2016), *Cuminum cyminum* can impart antimicrobial, anti-inflammatory, anticancer, insecticidal, antioxidant, analgesic, antiplatelet aggregation, antidiabetic, hypotensive, contraceptive, bronchodilatory, immunological, anti-amyloidogenic, aldose reductase, anti-osteoporotic, alpha-glucosidase and tyrosinase inhibitory effects, protective and central nervous effects.

Dubey *et al.* (2017) reported that cumin is one of the most important pharmaceutical, industrial and economical crops belonging to *Apiaceae* family and is particularly valuable for its culinary, nutritional and medicinal properties. For farmers in the arid and semi-arid regions of India, Iran, Syria, Pakistan and Turkey, cumin is a source of income, as a cash crop.

2.2 Cumin Oil

Li and Jiang (2004) isolated the essential oil of the seeds of *Cuminum cyminum L.* of China by hydrodistillation method and they examined the chemical composition of the oil. They identified 37 components like cuminal (36.31%), cuminic alcohol (16.92%), γ -terpinene (11.14%), safranal (10.87%), p-cymene (9.85%) and β -pinene (7.75%).

The physicochemical properties of the cumin seed essential oil may be affected by various factors such as plant variety, cultivation area and conditions, date of harvesting and extraction methods (Gohari and Saeidnia, 2011).

The flavour compounds of the oil, its quantity and composition primarily establishes the characteristic pungent flavour of cumin. Composition of the volatile oil determines the odour and flavour character of the spice. The yield of oil depends greatly upon the age of the seed; the older the seed lesser the volatile oil it contains (Ravi *et al.*, 2013).

Moghadam (2015) investigated the chemical composition of the hydro-distilled essential oil of *Cuminum cyminum L.* obtained from Iran by GC/MS. The various organic compounds found in the essential oils, were thymol (40.65%), γ-terpinene (24.51%), b-pinene (5.38%), a-pinene (3.47%), camphene (2.31%), terpinene- 4 – ol (2.00%), cuminaldehyde (1.79%), a-thujene (1.45%), a-terpinolene (1.17%), myrcene (1.07%), limonene (1.04%), α-phyllanderene (0.94%), acetoxylinalool (0.57%) and sabinene (0.37%).

Dubey *et al.* (2017) analysed the chemical composition of cumin essential oil obtained from Rajasthan and Gujarat states in India and reported that cuminaldehyde was the major constituent which varied from 25.84% to 39.90%. They concluded cuminaldehyde and γ –terpinene are the main chemical components present in Indian cumin oil.

2.3 Encapsulation

The encapsulation techniques are having increased applications in the food processing industry as it protects the encapsulated active compounds from heat, moisture or other extreme conditions and consequently enhance their stability and maintains viability (Gibbs *et al.*, 1999).

According to Madene *et al.* (2006) encapsulation is a process in which one material or a mixture of materials are coated with or entrapped within another material or system.

The encapsulated substance is known as active agent, core material, payload phase, or internal phase. The substance which is used for encapsulating or coating the core material is known as coating material, wall material, carrier material, membrane, shell, external phase or matrix. Reservoir type and matrix type are the two main types of encapsulates. In reservoir type encapsulate the active compound form a core and an inert

barrier surrounds the core. It is also known as mono-core, single-core or core-shell type. In case of matrix type encapsulates, the sensitive compound is dissolved or dispersed in an inert polymer. A combination of the first two is known as coated matrix type (Jeyakumari *et al.*, 2016).

Paredes *et al.* (2016) stated that particle encapsulation is a standard process used in the food processing industries which involve the encapsulation of materials in a protective layer, covering, or containment material that can protect a sensitive ingredient or nucleus from adverse reactions. The active core materials are encapsulated in the protective shells to preserve physico-chemical and organoleptic properties of the original products and also to enhance the palatability of volatile odorous ingredients.

The various types of encapsulation methods used include spray cooling or spray chilling, spray drying, extrusion coating, fluidised bed coating, coacervation, centrifugal extrusion, liposome entrapment, rotational suspension separation and inclusion complexation (Gibbs *et al.*, 1999).

Usually the encapsulation methods are based on drying processes because of the liquid nature of the extracts containing the bioactive components (Ballesteros *et al.*, 2017).

2.3.1 Microencapsulation

According to Rosenberg *et al.* (1985), microencapsulation is a process of packing small droplets of liquid or solid particles into continuous individual shells.

Bhandari *et al.* (1992) stated that microencapsulation process has become an attractive method for transforming liquid food flavourings into free flowing and stable powders which are easy to handle and can be

incorporated into a dry food system. The diluted state is also more convenient for mixing the flavouring agents with solid foods uniformly.

Rosenberg and Sheu (1996) stated that microencapsulation is a technique where a protective film or wall system is formed around the particles or droplets of the core or microencapsulated material. The wall systems or protective films are designed for protecting a sensitive core substance from those factors which can deteriorate them, to limit loss of volatiles, and also to prevent a premature interaction between the core or active material and other ingredients.

In food industry, the microencapsulation process is applied due to various reasons and are summarised by Desai and Park (2005) as follows: (a) encapsulation protects the core material from degradation by reducing its reactivity to its surrounding environment (e.g., moisture, heat, light and air) (b) retardation of transfer or evaporation rate of the active substances to the surrounding atmosphere (c) the product can be tailor to either release slowly over time or at a certain point (d) modification of physical properties of the original material and allows easier handling (e) masking of the flavour of the core compound (f) dilution of the core material when only very small amounts are required, yet still achieve a uniform dispersion in the host material and (7) can be used for the separation of components within a mixture that would otherwise react with one another.

Microencapsulation is the technology of enclosing solids, liquids, or gaseous substances in miniature, sealed capsules and under specific conditions they will release their contents at controlled rates. In encapsulation process, enzymes, cells, food ingredients or other materials in small capsules can be incorporated. (Lopez-Rubio *et al.*, 2006).

Nazzaro *et al.* (2012) stated that microencapsulation is a process of envelopment of small solid particles, liquid droplets or gases in a coating (1–1000 μm).

2.3.2 Nanoencapsulation

According to Yurdugul and Mozafari (2004), nanoencapsulation is a process that involves the incorporation of bioactive agents, including food ingredients, antioxidants, vitamins, slimming agents and enzymes, in small capsules with sub-micron size diameters. The application of nanoencapsulation in the food industry is getting increased due to various advantages provided by this method which include enhanced stability of the encapsulated material by protecting them from adverse conditions such as heat, humidity, pH variations and other extreme conditions as well as it masks unwanted odours or tastes.

In comparison with micron-sized carriers, nanocarriers can provide larger surface area, enhance the solubility, enhance controlled release, increase bioavailability and enable precision targeting of the encapsulated substances to a greater extent. The amount of materials required for exerting a specific effect is much less when encapsulated inside the nanocarriers than when unencapsulated due to the improved stability and targeting. Time-controlled and targeted release can improve the effectiveness of bioactives, broaden their applications range and ensure optimal dosage and hence improving the cost-effectiveness of the product. A nanocarrier system like nanoliposomes can be used to encapsulate a reactive or sensitive material for making them stable ingredients such as nanoliposomes (Mozafari *et al.*, 2006; Mozafari *et al.*, 2008)

Ezhilarasi *et al.* (2013) reported that by reducing the size of particles into nanoscale range, the surface-to-volume ratio can be increased, which thereby enhances their reactivity many folds with change in electrical, optical and mechanical properties.

2.4 Core material

Venkatesan *et al.* (2009) stated that the core material to be encapsulated can be liquid or solid. That means, if it is liquid it should be dispersed and/or dissolved material whereas the solid material should be a mixture of active constituents, stabilisers, diluents, excipients and release-rate accelerators or retardants.

2.5 Wall materials

For a good carrier material, the most critical criterion is its ability to seal and hold the active substance within its structure during processing and storage. Excessive loss of core material will take place when the carrier material is unable to meet the above criterion (Balassa and Fanger, 1971; Dian *et al.*, 1996). This will also lead to the presence of increased amount of oil/fat on the surface of the powder particles. This surface oil or unencapsulated oil is liable to be subjected to the action of atmospheric oxygen. These factors then affect other characteristics of the powder particles.

The main functions of the wall materials are to protect the core material (material which is encapsulated) from various factors that cause the deterioration of the core material such as oxygen, moisture and light. It also permits the controlled release of the active material under desired conditions (Rosenberg *et al.*, 1985).

The wall materials used for protecting the core material by encapsulation and its controlled release can significantly impact the retention of the core substances and also the performance and functionality of the final product. It is especially true in the case of flavour encapsulation (Risch, 1995).

Desai and Park (2005) has summarised the important characteristics that should be possessed by an ideal coating material which are: (i) The ability to seal and hold the core compound within its structure during processing or storage, (ii) Good rheological properties at high concentration and easy workability during encapsulation, (iii) Nonreactivity with the encapsulated material during processing and storage, (iv) The ability to disperse or emulsify the core substance and stabilising prepared emulsion, (v) The ability to protect the sensitive core substance from adverse environmental conditions (e.g., light, humidity, heat, oxygen), (vi) Solubility in solvents acceptable in the food industry (e.g., ethanol, water), (vii) The ability to completely release the solvent or other materials used during encapsulation process under drying or other desolventisation conditions, (viii) Chemical nonreactivity with the core compounds, (ix) Low cost and food grade.

Drusch *et al.* (2007) classified the wall materials used for encapsulation into gums (agar agar, gum arabic, locust bean gum), lipids (palm fat, wax), proteins (gelatin, milk proteins, soy protein), polysaccharides (alginate, guar gum, pullulan, starch, xanthan), and mono-, di-, and oligosaccharides (hydrolysed starch, lactose) as well as cellulose and its derivatives (carboxymethylcellulose, methylcellulose).

Bansode *et al.* (2010) has listed the various properties that should be possessed by the coating materials used for the encapsulation process as follows: (a) stabilisation of active core material, (b) nonreactive towards the encapsulated active component, (c) controlled release of the active component under specific conditions, (d) soluble in an aqueous media or solvent or melting (e) low viscosity, non-hygroscopic and economical, (f) stable, film-forming, tasteless and pliable, (g) the coating can be flexible, thin, brittle, hard etc.

Nedovic *et al.* (2011) reported the most important criteria for the selection of suitable wall materials are; functionality that the encapsulate or wall materials should provide to the final product, potential restrictions for the coating material, type of release, concentration of encapsulates, cost constrains and stability requirements. It is important that the materials used as the encapsulate or protective shell must be food-grade, biodegradable and also capable of forming a barrier between the internal phase and its surroundings. In the food sector, biomolecules forms the majority of the materials used for encapsulation purpose.

Various materials used for encapsulating agents include carboxy methyl cellulose, chitosan, starches, gelatin, guar gum, gum arabic, maltodextrins, sodium alginate, sodium caseinate, pectin etc. (Ravichandran *et al.*, 2014). However according to Fathi *et al.* (2014), carbohydrate-, lipid- or protein- based materials are the most suitable nano-scale coating agents for food applications.

2.5.1 Gum Arabic

Several researchers have investigated the use of gum arabic and maltodextrin as coating materials -unmixed and mixed- for encapsulating different bioactive materials.

According to Street and Anderson (1983), apart from the widespread uses of gum arabic as a foodstuffs additive, it is one of the important ingredients in confectionery items which can create various textures and other specialised effects.

Though gum arabic was known as excellent emulsifier used in spray drying, it has limited application in food industry since the use gum arabic is fairly expensive when compared to maltodextrins (Shiga *et al.*, 2001).

Turchiuli *et al.* (2005) stated that acacia gum which is a hydrocolloid produced by natural exudation of acacia trees is composed of a highly branched arrangement of the simple sugars (galactose, arabinose, rhamnose) and glucuronic acids. A protein component (~2% w/w) is present in them which is covalently bound within its molecular arrangement and this component have a vital role in determining their functional properties like high solubility in water, bland flavour, low viscosity of concentrated solutions compared to other hydrocolloid gums and very good oil-in-water emulsifier.

Gum arabic is a gum that is most commonly used as a flavour-encapsulating material in spray drying process. Its low viscosity in aqueous solutions, solubility, good emulsification properties and its good volatile retention make it very versatile for most of the encapsulation techniques. As a carrier material, gum arabic is ideally suited for lipid droplets encapsulation. It can acts as both surface-active agent and drying matrix, thereby prevents the loss of volatiles that comes in contact with the atmosphere (Madene *et al.*, 2006). They also reported that the typical size of spray dried powder particles produced by combinations of maltodextrin and gum arabic are 10–200 μm. The volatile component retention, which is usually >80%, depends on various parameters which include inlet temperature of the spray dryer, viscosity and concentration of the emulsion and gum arabic to maltodextrin proportion.

Kanakdande *et al.* (2007) stated that gum arabic can produce a stable emulsion with most of the oils over wide pH range and can also form a visible film at the oil interface. Due to high cost, variations in the quality and limited supply, the use of gum arabic in the encapsulation purpose is limited.

Ferrari *et al.* (2013) evaluated the stability of anthocyanins and antioxidant activity of blackberry powder, produced by spray drying

method, with maltodextrin, gum arabic, or a blend of both as wall materials. Their results showed that powder produced with maltodextrin as carrier agent had the longest half-life and the lowest degradation rate of anthocyanin at 25°C. Their results showed that maltodextrin offered greater stability for spray dried blackberry powder. They concluded that the use of maltodextrin or the combination of both maltodextrin and gum arabic as wall materials could promote better maintenance of antioxidant potential of blackberry powders.

Alves *et al.* (2014) stated that the colloid functionality of gum arabic makes it an effective coating material in the encapsulation processes. It is compatible with most gums, carbohydrates, proteins and starches. Maltodextrin, a hydrolysed starch are commonly used as a carrier agents due to their low price, high water solubility, good flavour and the low viscosity of the solutions it forms. Hence mixtures of gum arabic and maltodextrin can provide a good balance between efficiency and cost.

Acacia gum or gum arabic is the exudate obtained from the plants, Acacia seyal and Acacia senegal, belonging to the *Leguminosae* family. It is a complex, branched heteropolysaccharide, either neutral or slightly acidic (Patel and Goyal 2015).

2.5.2 Maltodextrin

Desobry *et al.* (1997) stated that the most commonly used coating materials are maltodextrins due to their good efficacy and reduced cost. They are bland in flavour and possess low viscosity at higher solids ratio. These are available in different average molecular weight fractions (DE values) of 4, 10, 15, 20, 25, 30 and 42 and as DE increases, the average molecular weight decreases. Hence blending can create different wall densities.

Cai and Corke (2000) reported that the usage of maltodextrin as the coating or carrier agent on the production of spray dried *Amaranthus* Betacyanin pigments considerably reduced the hygroscopicity of the betacyanin extracts.

According to Carneiro *et al.* (2013), maltodextrins have relatively lower cost, neutral taste and aroma, reduced viscosity at increased solids concentrations and good protection against oxidation.

Maltodextrin is a polysaccharide produced from starch by its partial enzymatic hydrolysis. They have a dextrose equivalence less than 20 which indicates that it has long carbohydrate chains along with 2-3% glucose and 5-7% maltose and is available in white hygroscopic spray-dried powder which is slightly sweet almost flavourless. Maltodextrin is considered as good encapsulant since it exhibits low viscosity at high solids contents and also good water solubility. Additionally, the gelling properties of polysaccharides can stabilise the emulsion towards flocculation and coalescence. As maltodextrins can be dispersed in water up to 35.5% of the solution without the formation of haze, these materials can contribute highest flavour retention (Parikh *et al.*, 2014).

Mahdavi *et al.* (2016) reported that a combination of maltodextrin and gum arabic as carrier agents offered highest process efficiency and best powder quality in the microencapsulation of natural anthocyanins.

2.5.3 Tapioca Starch

Loksuwan (2007) conducted a study on microencapsulation of b-carotene by spray drying with modified tapioca starch, native tapioca starch and maltodextrin as the wall materials. They observed that the modified tapioca starch exhibited wider particle size distribution, towards the smaller diameters compared to maltodextrin and native starch. They also observed

that the highest level of total b-carotene was possessed by modified tapioca starch. Modified tapioca starch had lowest surface b-carotene.

Setyaningsih *et al.* (2009) studied the microencapsulation of natural vanilla extract by spray drying technique with maltodextrin DE10 and modified cassava starch (Flomax 8) as encapsulant. Their results showed that vanillin extract encapsulated by maltodextrin DE10:Flomax8 2:1 gave higher yield, vanillin content and vanillin recovery than only maltodextrin. A 30% concentration of coating agents gave the highest vanillin content and vanillin recovery. Higher vanillin content was obtained for higher ratio of vanillin extract to coating material, but resulted in lower recovery.

Ordonez and Herrera (2014) encapsulated limonene by spray drying with the wall materials of gum arabic, whey protein concentrate, cassava starch and gum arabic and cassava starch and whey protein. Their results showed that, mixtures of cassava starch–gum arabic and whey protein–cassava starch allowed limonene to be captured in proportions above 40% by spray drying process. They observed capsule morphologies showed no cracks and had uniform surfaces, which indicated that the wall materials and the encapsulation process gave sufficient protection to limonene.

2.6 Spray Drying

In a spray drying operation, atomisation is the most important step as it determines the size of final products, and the initial drop size distribution of the spray generated by the atomiser forms the basis of the chamber design (Gauvin and Katta, 1976).

Newton (1966) cited by Broadhead *et al.* (1992) stated that spray dried powders are usually spherical and hollow with a narrow size distribution. The hollow nature provides lower bulk density to the powder particles, but despite this, their spherical shape means that they are usually free-flowing. They also suggested that by the modification of the spray

drying technique, the particle size and its distribution, appearance, moisture content, particle density, bulk density, flowability, porosity, stability, friability, dispersability and retention of activity, aroma and flavour can be altered and controlled. The particle properties are affected by design of the nozzle and drying chamber.

Shahidi and Han (1993) reported that spray drying is the most commonly used encapsulation method in the food industry. This process is flexible and economical, and can produce good quality powders. Usually this technique is considered as a dehydration process, but it can also be used as an encapsulation method when it entraps "active" compound within a protective matrix formed from a polymer or melt.

According to Desobry *et al.* (1997), the cost of spray-drying method is 30–50 times cheaper when compared to freeze-drying. They also reported that the simplest means of encapsulation is to emulsify the sensitive ingredient (usually oil) in a wall or coating material solution followed by drying such that the coating agent then entraps or coats the sensitive ingredient within it and provides barrier to water vapour and oxygen.

According to Re (1998), atomiser selection is one of the very important factor in the design of a spray dryer which significantly affect the size distribution of the final dried particles. Uniformity of drop-size, its distribution, and homogeneity of the spray are the most important characteristics of the atomiser from the standpoint of product quality. Common forms of atomisers are pneumatic (two-fluid) atomisers, centrifugal (wheel) atomisers and pressure atomisers.

Spray drying is a unit operation by which a liquid product is atomised in a hot gas current to instantaneously obtain a powder and the initial liquid feeding in to the spray dryer can be an emulsion, a solution or a suspension (Gharsallaoui *et al.*, 2007). Particle size of the final product obtained from the spray dryer is mainly dependent on the feed material and

operating conditions. Based on this, spray drying can produce a very fine powder (10–50 μm) or large size particles (2–3 mm).

Fernandes *et al.* (2008a) reported that spray drying is one of the most widely used techniques for the production of solid encapsulated forms of essential oils. Generally, it can provide better protection to the core material and thereby increase the shelf life of the powder products.

Tonon *et al.* (2008) studied the influence of process conditions on the physicochemical properties of acai powder produced by spray drying. They used maltodextrin 10DE as carrier agent. Independent variables chosen were: inlet air temperature (138–202°C), feed flow rate (5–25 g/min) and maltodextrin concentration (10–30%). By increasing the spray dryer inlet temperature, higher process yield and powder hygroscopicity, lower moisture content and anthocyanin retention were observed. Feed flow rate showed negative influence on hygroscopicity and process yield, and positive influence on moisture content. Concentration of maltodextrin negatively affected the powder hygroscopicity, which confirmed its efficiency as a coating material. Increasing maltodextrin concentration resulted in a reduction of process yield. Increasing temperatures resulted in the production of larger particles and most of them had smooth surface.

Adamiec (2009) reported that the two main limitations of spray drying technique are: (1) the core material should be heat resistant to the temperature of hot air usually applied in this drying method even at short exposure time to hot air, (2) limited number of coating agents, which should be film-forming and should have an acceptable level of water solubility.

Among the various techniques used for the encapsulation of food materials such as physical (air suspension coating, centrifugal extrusion, pan coating, spray drying, vibrational nozzle, etc.) and chemical (interfacial polymerisation, in-situ polymerisation, matrix polymerisation, etc.) methods, spray drying is the most commonly used method due to its low

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operational cost and ability to handle labile materials (Aghbashlo *et al.*, 2012).

In general, the process of spray drying consists of four steps (Manu *et al.*, 2012). The first stage is atomisation of the liquid feed into a spray of droplets. The second stage involves the contact of spray-air, mixing and droplet/particle flow. In the third stage, drying and particle formation take place. When the spray droplets come in contact with the drying air, the solvent evaporation takes place. In the last stage, separation of particles from the drying air and discharge of the dried products to the drying chamber and associated particle collection systems (cyclone, filter bag, scrubber) takes place.

The nozzle of the spray dryer makes the sprayed droplets as small as possible for maximising the heat transfer and rate of water vaporisation. In a spray dryer, the drying of the feed take place very rapidly compared to other drying techniques. This technique has the advantage of maximising the profit and minimising the process as it can convert a slurry or solution into dried powdered products in a single step (Phisut, 2012).

Sosnik and Seremeta (2015) reported that spray drying is a rapid, cost-effective, continuous, scalable and reproducible technique for producing dried powders from a fluid material by atomisation. Spray drying can be adopted for encapsulating hydrophilic and hydrophobic materials within various coating materials without significant thermal degradation, even of heat-sensitive materials because of rapid drying (seconds or milliseconds) and relatively short exposure time to heat. The solid particles produced have relatively narrow size distribution at the submicron-to-micron scale.

The volatile losses during the drying process mainly occur from three regions as described by King (1995) and Jafari *et al.* (2008). First is atomisation during which major volatile loses can take place due to the

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substantial surface areas, turbulence and secondary flows within the sheets and ligaments of liquid that are drop precursors. Second is from undisturbed drops. Once droplet circulation ceases due to surface drying and/or forces from surface-tension gradients, volatile losses take place by diffusion within the liquid phase. The final volatile loss takes place during the morphological development. Morphological development during periods of expansion and cratering of drops exposes surfaces that can lose volatile components, but the concentration of the drop solutions will also takes place when this happens.

2.7 Encapsulation Using Spray Drying

Krishnan *et al.* (2005) conducted a study on the microencapsulation of cardamom oleoresin by spray drying. The wall materials used were binary and ternary blends of gum arabic, maltodextrin, and modified starch. They evaluated the content and stability of volatiles, entrapped 1,8-cineole and entrapped a-terpinyl acetate for 6 weeks. They concluded gum arabic as a better carrier material for encapsulating cardamom oleoresin compared to others. In case of blends, they observed a decrease in the stability of the cardamom oleoresins when the proportion of gum arabic was decreased in its blend with maltodextrin and modified starch.

Adamiec and Kalemba (2006) discussed the results of preliminary investigations of the preservation of elemi and peppermint oils in a maltodextrin microcapsule during spray drying. Their results confirmed that the spray drying technique can be applied in the microencapsulation of multicomponent emulsions of essential oils in a coating agent. When the boiling points of encapsulated substances were higher, the efficiency of the process was higher.

Kanakdande *et al.* (2007) conducted a study on microencapsulation of cumin oleoresin by spray drying method. The wall materials used were gum arabic, maltodextrin, and modified starch (HiCap® 100) and their

ternary blends. They concluded gum arabic as a better wall material for encapsulation of cumin oleoresin than other wall materials. Their results showed thst a 4/6:1/6:1/6 blend of gum arabic:maltodextrin:modified starch as more efficient than other combinations, and even better than gum arabic itself.

Bae and Lee (2008) stated that the preparation of a stable emulsion with proper chemical and physical properties is one of the main prerequisites in the microencapsulation of oil by spray drying process. The emulsion stability, properties and composition determine the quality characteristics of the spray dried oil powder like microencapsulation efficiency, structure, surface free oil content, physical properties (bulk density, dispersibility and flowability) and oxidative stability.

Laohasongkram *et al.* (2011) conducted a study on the microencapsulation of macadamia oil by spray drying technique. The optimum conditions for emulsion preparation were studied by changing the carrier material to core material ratio, sodium caseinate to maltodextrin ratio, and homogenizing pressure. They concluded that the optimum conditions for microencapsulation of macadama oil as a sodium caseinate:maltodextrin ratio of 1:4, wall material:core material ratio of 60:40 and 200 bar homogenizing pressure. A feed rate of 1.1 kg/h and 167°C spray dryer inlet temperature were the optimum conditions of spray drying operation.

Rocha *et al.* (2012) microencapsulated lycopene by spray drying method, with a modified starch as coating material. Encapsulation efficiency varied between 21 and 29% and they observed that the microcapsules possessed a rounded outer surface with the formation of concavities and they varied in size. They conclude that the microencapsulation provided greater protection to lycopene compared to its free form.

Alves *et al.* (2014) conducted a study on the microencapsulation of the essential oil from the fruits of *Pterodon emarginatus* with spray drying technique. They used gum arabic and maltodextrin as the wall materials. From the X-ray diffraction, it was observed that the essential oil was entrapped inside the capsules rather than being adsorbed onto the particle surfaces. The best protection for the oil was offered by the powder produced from a 1:3:3.6 blend of essential oil: gum arabic: maltodextrin. With this blend, 98.63% of the oil was retained inside the microcapsule and the same proportion of b-caryophyllene was entrapped.

Fathi *et al.* (2014) reported that in an encapsulation process, the various factors on which the encapsulation efficiency and the particle size depend include the properties of the materials used, e.g. properties of core and wall materials, viscosity of the solution, particle size (for emulsified active ingredients), spray dryer factors like type of atomiser, inlet and outlet temperatures, flow rates and humidity. As the feed flow rate decreases, the particle size decreases and the encapsulation efficiency increases. But with the increase in inlet temperature, particle size and encapsulation efficiency increases because of the hard crust formation. The particle size increases also with increased air flow rate. Higher encapsulation efficiency may be obtained with a moderate air flow rate. The efficiency and the particle size decreases with the decrease in humidity.

Al-Ismail *et al.* (2015) conducted a study on microencapsulation of cardamom's essential oil (CEO) with Gum Arabic (GA), whey protein isolate (WPI) or their combinations (WPI+GA) at different mixing proportions as the carrier substances by spray drying method. Their results showed that GA microcapsules had the highest encapsulation efficiency and retention of CEO throughout the storage period regardless of the storage temperature. WPI microcapsules showed the lowest encapsulation efficiency and CEO retention. Both WPI+GA combinations had intermediate microencapsulation efficiency and retention of CEO compared

to the single component matrices. Both WPI and GA microcapsules had spherical shape. They concluded that GA microcapsules showed the best overall encapsulation properties than others.

Kausadikar *et al.* (2015) conducted a study on the microencapsulation of lemon oil with three carrier materials *viz*,. gum arabic, maltodextrin, modified starch and their binary and ternary blends by spray drying process. The parameters such as carrier concentration (30%), core material concentration (10%), inlet temperature (175°C) and outlet temperature were found to be effective for maximum encapsulation efficiency. Gum arabic gave maximum encapsulation efficiency than others due to large particle size and it also gave minimum surface oil concentration but maximum total oil concentration which is a great requirement of any carrier material. They concluded that gum arabic:maltodextrin at 50:50 combination provided the highest encapsulation efficiency (82.60%).

Kalal *et al.* (2016) conducted a study on the production of nanoencapsulated probiotic bitter gourd juice powder using spray dryer at different inlet air temperatures. Maltodextrin and gum arabic were used as the coating materials. Their results showed that the increase in the spray dryer inlet temperature reduced the moisture content, water activity and bulk density of the powder while increased the solubility. Water activity, moisture content, solubility and bulk density were significantly affected also by the maltodextrin concentration.

Cortes-Camargo *et al.* (2017) microencapsulated lemon essential oil (LEO) with Mesquite gum (MG) and nopal mucilage (NM) mixtures as wall materials by spray drying. Higher concentrations of NM in emulsions showed larger droplet sizes and higher viscosity values and their microcapsules also exhibited larger particle sizes with the highest values of encapsulation efficiency and oxidative stability probably because of the more extended mucilage structure that allows a better coverage of the oil

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surface droplets, besides to enhance the viscous, steric and electrostatic effects. They proposed MG–NM mixtures as new wall materials as they could contribute to improve certain desirable characteristics such as a high retention and shelf life extension of LEO in microcapsules regarding using biopolymers separately.

Ogrodowska *et al.* (2017) investigated the influence of drying process conditions on the physical properties, bioactive compounds and stability of encapsulated pumpkin seed oil. Their results showed that the bulk density decreased with the increase in the spray dryer inlet temperature. More amount of free oil was generated with increased spray dryer inlet temperature. At 130°C inlet temperature they got highest encapsulation efficiency in the spray drying process. But at this 130°C inlet temperature, deformation of the capsules was more than those produced at higher temperatures.

2.8 Response surface methodology

According to Bas and Boyacı (2007), response surface methodology (RSM) consists of a collection of statistical and mathematical techniques that can be used for defining the relationship between the independent variables and responses. It can define the effect of independent variables, alone or in combination, on the processes.

According to Ferreira *et al.* (2007), the Box-Behnken design is a good design for response surface methodology because it permits: (i) estimation of the parameters of the quadratic model; (ii) building of sequential designs; (iii) detection of lack of fit of the model; and (iv) use of blocks.

Ahn *et al.* (2008) optimised the microencapsulation condition of sunflower oil by employing the response surface methodology (RSM). They investigated the microencapsulation efficiency of encapsulated sunflower oil

with respect to four parameters such as sunflower oil concentration, proportion of milk protein isolates to coating wall, soy lecithin concentration, and homogenizing pressure. The optimal conditions obtained were 23.6% sunflower oil, 19.0% milk protein isolates, 2.5% soy lecithin, and 54.8% dextrin.

Application of RSM for the optimisation of analytical procedures is largely diffused and consolidated today due to its advantages to classical one-variable-a-time optimisation. It can generate large amounts of information from few numbers of experiments and can evaluate the interaction effect between the variables on the responses (Bezerra *et al.*, 2008).

Response Surface Methodology is a powerful statistical procedure which is frequently used in many engineering applications to build accurate models in an optimisation design (Aghbashlo *et al.*, 2012).

RSM is used for developing and improving optimisation of process parameters. It is most extensively applied in various stages like determination of multi-response parameters and their effective levels, experimental design selection, prediction and verification of model equations, generating response surfaces, contour plots and determination of optimum conditions. With minimum cost and time, RSM can be used for obtaining high efficiency for the development of improved or new process or products (Balasubramani *et al.*, 2015).

2.9 Qualities characteristics of the encapsulated powders

Kieckbusch and King (1980) investigated the losses of volatile acetates in the vicinity of pressure nozzles during spraying of sucrose and maltodextrin solutions. Their results showed that volatile loses are very high in the immediate vicinity of the nozzle, where only small amount of water has evaporated.

According to Rosenberg *et al.* (1988), the protection offered by the coating materials and the flow characteristics of the encapsulated products depend on the inner and outer microstructure of the encapsulated capsule and also on how the active compound is organized within the capsule. The porosity, bulk density and retention of the volatile core substance are determined by the microstructure of the spray dried products.

Rosenberg and Sheu (1996) conducted a study on microencapsulation of volatile esters with whey protein isolate or a mixture of whey protein isolate and lactose as the wall materials. Their results showed high retention of volatiles. With increased wall solids concentration, the retention was increased whereas the initial volatile load showed adverse effect on the retention. By incorporating lactose with whey protein isolate, the volatile retention was improved during the drying process and it limited the extractability of the core compound.

Re (1998) stated that the volatile retention in the spray drying process can be enhanced if increased dissolved solids concentration is built up on the droplet surface early enough in the drying process. Hence, the temperature and concentration of feed are very important because of their effect on the crust formation. Increased volatile retention can be obtained with hotter air due to faster drying which imposes steeper internal water concentration gradients, but this must be balanced against the possibility of greater thermal degradation.

According to Gibbs *et al.* (1999), there are different forms of encapsulation like a simple membrane coating, spherical or irregular shaped wall or membrane, multiwall structures having walls of the same or varying compositions or numerous cores within the same walled structure.

Apintanapong and Noomhorm, (2003) conducted a study on the microencapsulation of 2-acetyl-1-pyrroline, a major flavour component of aromatic rice by spray drying technique. For finding out proper coating

agents for microencapsulation of 2-acetyl-1-pyrroline, they used different ratios of gum acacia and maltodextrins. Encapsulation with a gum arabic:maltodextrin ratio of 70:30 offered the best retention of 2-acetyl-1-pyrroline. Their study revealed that the presence of higher maltodextrin ratios in powders resulted in lower moisture content.

Depending on the encapsulation process used, the encapsulation matrices may have different shapes (spheres, films, irregular particles), different structures (compact or porous) and different physical structures (amorphous or crystalline dehydrated solid, glassy or rubbery matrix) that can affect the diffusion of external substances (oxygen, solvent) or flavours and also the stability of the products during storage (Madene *et al.,* 2006).

In case of oil encapsulation, the encapsulation efficiency is considered as one of the most important quality characteristics of the powders. It is defined as the percentage of encapsulated oil in total oil. The encapsulation efficiency is usually calculated by determining the surface or unencapsulated or free oil which is present on the surface of the encapsulated capsules by washing the encapsulated powders with organic solvents. This surface oil adversely influences the physical properties of the encapsulated powders like flowability, bulk density and dispersibility. Also it can induce more rapid oxidation of the lipids. But the analysis of unencapsulated oil by solvent extraction method may also accounts for the encapsulated oil extraction to some extent since the solvent can reach the interior through cracks and pores. Hence the value of free oil content goes higher than the actual value (Velasco *et al*., 2003; Vega and Roos, 2006; Bae and Lee, 2008).

Bae and Lee (2008) conducted a study on the microencapsulation of cold pressed avocado oil with four different coating materials including whey protein isolate alone or in combination with maltodextrin (DE 5) by spray drying process. Their results showed that increased maltodextrin

proportion in the wall system improved wettability and density of the powders.

Jafari *et al.* (2008) reported that a successful spray drying encapsulation is achieved when the volatile retention is high and the amount of surface oil present on the encapsulated powder particles is minimum for volatile and non- volatile materials during the process and also under storage periods. The main factors influencing the retention and efficiency of the core material are the characteristics of both core and coating agents and the emulsion prepared for the spray drying conditions. The core-to-wall ratio represents the flavour or oil concentration or the load. Usually an optimum core concentration is there which can be efficiently encapsulated. Higher oil loads will usually result in poorer retention or lower encapsulation efficiency and higher surface oil content of the encapsulated powder.

According to Dubey *et al.* (2009), the microscopic size is the most significant charecteristics of microcapsules which provides a large surface area. One millimetre of hollow microcapsules having a diameter of 0.1 mm has total surface area of about 60 m^2. The total surface area and the diameter are inversely proportional. These large surface areas are available for sites of adsorption and desorption, light scattering, chemical reactions, etc.

Janiszewska and Witrowa-Rajchert (2009) categorised the wall materials used in the encapsulation process as agglomerate, skin-forming and crystalline structures. Usually a round or an oval shape with varied surface structures are expected depending on the type of material applied. In the food industry, skin-forming type materials are most frequently employed and the particles of such substances (e.g. maltodextrin, skim-milk) have smooth surface, where craters, holes, cavities, air vacuoles and wrinkling may be present directly below the particle surface. They also reported that increased particle size may be obtained when aroma is encapsulated in this

type of skin-forming materials at increased temperatures due to the 'ballooning and puffing' processes taking place during spray drying.

Toure *et al.* (2011) microencapsulated volatile ginger oil by spray drying technique. Maltodextrin and whey protein isolate were chosen as the coating substances. They concluded that the best conditions for higher microencapsulation efficiency and oil retention were a whey protein isolates to maltodextrin ratio of 1:1, a core to wall material ratio of 1:4, and 25% total solid content. Microencapsulation efficiency and oil retention were 99.36% and 93.30%, respectively. SEM analysis revealed that no visible cracks or pores were present on the microcapsule.

Fernandes *et al.* (2013) reported that the bulk density, particle density and respective particle porosity, moisture content, and the instantanisation properties (wetting, dispersibility, and solubility) of the microencapsulated essential oils are the important physical properties in relation to their ease of dispersion in aqueous solutions. These physical properties are mainly affected by the characteristics of the feed (solids content, temperature and viscosity), type of spray dryer, operating pressure and speed and inlet and outlet air temperatures.

Krithika *et al.* (2014) conducted a study on microencapsulation of paprika oleoresin with 100000 CU with gum arabic as the carrier agent by the spray drying technique. Their results showed that the storage life of paprika oleoresin (5-15%) was maximised by microencapsulating it with gum arabic as the matrix material. From the scanning electron microscopy study, they observed that the powder particles microencapsulated with 10 and 15 per cent, possessed superficial indentations and dents like honey comb structure. The surface possessed lesser breakages and cracks, which provided increased protection to the paprika oleoresin.

The plasticizer characteristics, exhibited by the carbohydrate biopolymers can promote the development of smooth and spherical surface

microparticles and also increase the bonding strength between core and carrier substances (Campelo *et al.*, 2017).

Fernandes *et al.* (2017) conducted a study on the encapsulation of ginger essential oil by spray drying process and evaluated the effects of blending of whey protein isolate with inulin and maltodextrin as wall materials. They found that maltodextrin improved the encapsulation efficiency and inulin greatly improved the wettability and solubility of the encapsulated powder. They observed an increase in the thermal stability of the encapsulated ginger oil by all the coating material systems. Higher protection was offered by whey protein isolate: maltodextrin system with lower rates of degradation. Presence of inulin in powders resulted in lower water adsorption at high relative humidity.

Lee *et al.* (2017) conducted a study on the encapsulation of Asian pear juice by spray drying with different maltodextrin levels and inlet air temperatures. Their results showed that the L* and b* values of the encapsulated powders were significantly affected by the concentration of maltodextrin and the spray dryer inlet air temperatures. They observed that increase in the spray dryer inlet air temperature resulted in an increase in the total phenolic content while a reduction in vitamin C content. When higher inlet air temperature and maltodextrin concentrations were used, the encapsulated powders possessed lowest median particle diameter along with rounded, more regular and smoother outer surface. From their results they concluded that the addition of 15% (w/v) maltodextrin and spray drying at an inlet temperature of 170°C can produce very good quality encapsulated powder of Asian pear juice.

5H

Materials and Methods

CHAPTER III

MATERIALS AND METHODS

This chapter deals with the various materials and methods adopted for the production of both microencapsulated and nanoencapsulated cumin oil powders by spray drying technique in detail. It includes the different methods used for the evaluation of the quality parameters of the encapsulated powders and also the experimental procedures adopted for the optimisation of process variable on the basis of these quality parameters.

3.1. RAW MATERIALS

3.1.1. Core Material

Cumin (*Cuminum cyminum Linn.*) oil extracted by steam distillation procured from Vaidyaratnam P.S. Varier's Arya Vaidya Sala, Kottakkal, Kerala, India was used as the core material in microencapsulation and nanoencapsulation. The oil was packed well and kept out of sunlight and stored at room temperature.

3.1.2. Wall Material

Three wall materials or carrier materials *viz.,* gum arabic, maltodextrin and tapioca starch were used for the encapsulation of cumin oil. Gum arabic was procured from M/s. Chemind, Thrissur. Kerala. Maltodextrin with a dextrose equivalence of 20 (DE 20) was procured from M/s. Viveka agencies, Coimbatore and tapioca starch was obtained from CTCRI, Thiruvananthapuram, Kerala. All the chemicals and reagents used in this work were of analytical grade.

Preliminary studies were conducted with the wall materials (maltodextrin, gum arabic and tapioca starch) at various spray dryer inlet temperature for optimisation of suitable wall material or combination of

them and the spray dryer inlet temperature for the encapsulation of cumin oil. The preliminary studies included experiments with gum arabic, maltodextrin and tapioca starch individually as the carrier material and a binary blend of them at various inlet temperatures.

From the study it was observed that the tapioca starch was unable to significantly encapsulate the cumin oil by spray drying and hence it was eliminated from further studies. In case of experiments with individual carriers, gum arabic offered the best encapsulation efficiency for cumin oil than the others. But when gum arabic alone was used, the emulsion became highly viscous and it resulted in atomisation difficulties and semi-wet droplets were deposited on the wall of drying chamber of the spray dryer. The same result was obtained by Badee *et al.* (2012) in microencapsulation of peppermint oil by spray drying. They concluded that the concentration of gum arabic should not be increased beyond 30% for the microencapsulation of peppermint oil by spray drying, as higher gum concentration produces solution which is very viscous to be pumped through the atomiser of the spray drier. On the other hand, when maltodextrin alone was used, it produced sticky deposits of thermoplastic powder on the cyclone wall which was same as observed by Fernandes, *et al.* (2008b). But when binary blends of gum arabic and maltodextrin were used for the encapsulation, it showed better retention of the oil than that with gum arabic alone. Hence in this present work, binary blends of gum arabic and maltodextrin were selected as suitable wall material combination for both microencapsulation and nanoencapsulation of cumin oil by spray drying.

3.2. PHYSICO-CHEMICAL PROPERTIES OF RAW MATERIALS

3.2.1 Physico-chemical Properties of Cumin Oil

3.2.1.1 Colour

A Hunter lab colour flex meter (Hunter Association laboratory, Inc., Reston, Virgina, USA; model: HunterLab's ColourFlex EZ) shown in Plate 3.1 was used to find out the colour values of cumin oil. The Hunter lab's colour flex spectro calorimeter consists of measurement (sample) port, opaque cover and display unit. This colour flex meter works on the principle of focusing the light and measuring energy reflected from the sample across the entire visible spectrum. For matching a series of colour across the visible spectrum, primary lights are required and describes the colour by mathematical model called as Hunter model. It reads the colour of sample in terms of L*, a* and b* values where, luminance (L) forms the vertical axis, which indicates whiteness to darkness. Chromatic portion of the solids is defined by: a (+) redness, a (-) greenness, b (+) yellowness, and b (-) blueness. A transparent glass cup filled with sample was placed over the port of the instrument and an opaque cover which act as a light trap to exclude the interference of external light was placed over the cup. Colour was calibrated by fixing the definite colours like white and black tiles. After calibration, the sample was placed over the port and values of L*, a* and b* were recorded.

3.2.1.2. pH

The pH of cumin oil was measured using a digital pH meter (M/s. Systronics; Model MK VI) which is shown in Plate 3.2. Initially the pH meter was standardised with double distilled water having a pH of 7.0 and standards of pH 4.0, 7.0 and 9.0. Then the pH of cumin oil was measured.

3.2.1.3 Specific gravity

Specific gravity of cumin oil was calculated by dividing the weight of 1 ml cumin oil by the weight of 1 ml distilled water. Weights were measured using a balance with an accuracy of 0.001g (Gopika and Ghuman, 2014).

3.2.2 Physico-chemical Properties of Wall Materials

3.2.2.1 Moisture Content

Moisture content of the wall materials (gum arabic and maltodextrin) was determined by the method AOAC (1999). Ten grams of accurately weighed wall materials were taken in petridishes and dried for 16 h in an electric oven at 70°C. Then the powders were taken out and cooled in a desiccator and finally weights were measured. The procedure was repeated until the differences between the two successive values of weights were not more than 0.5-1 mg. The initial and final values of weights were used to calculate the moisture content of the wall materials by the following equation and the value of moisture content was recorded.

$$\text{Moisture content (M) (\% w.b.)} = \frac{W_i - W_d}{W_i} \times 100 \qquad \text{.....(3.1)}$$

Where,

M = Moisture content, % (wet basis)

W_i = Initial weight of the powder, g

W_d = Dry weight of the powder, g

3.2.2.2 Water Activity

The water activity of wall materials were measured using Aqua lab water activity meter (model: Aqua lab, Decagon Devices Inc., Pullman

(Wa), USA) (Plate 3.3). For measuring the water activity of the each wall material, it was filled in the disposable cups of the water activity meter and turned the sample drawer knob to OPEN or LOAD position. Then the drawer was pulled to open. After the drawer gets opened the disposable cup along with the wall material was placed in the drawer. While placing in the drawer, check the top lip of the cup to ensure that it is free from sample residue (if the sample cup is over-filled, it may contaminate the sensors in the chamber). After that carefully slide the drawer closed. Then sample drawer knob was turned to the READ position for sealing the sample cup with the chamber and the water activity of the powder was shown in the LCD display of the water activity meter.

3.2.2.3 Bulk Density

The bulk density of the wall materials were determined according to the tapping method described by Beristain *et al.* (2001). In this method two grams of each wall material were loosely weighed into 10 ml graduate cylinder. The cylinder along with the powder was then tapped on a flat surface until a constant volume was achieved. The final volume was noted and bulk density was calculated by dividing the powder weight by the final volume of the powder in the cylinder as given below. The bulk density was expressed in g/cm^3.

$$\text{Bulk density}\,(g/cm^3) = \frac{\text{Weight of wall material}\,(g)}{\text{Volume of wall material}\,(cm^3)} \qquad .....(3.2)$$

3.2.2.4 Colour

The colour values of carrier materials were measured by using Hunter lab colour flex meter (Hunter Association laboratory, Inc., Reston, Virgina, USA) as described in the previous section 3.2.1.1. Samples were filled in the transparent glass cup and it was then placed over the port of the

instrument. The opaque cover was placed over the cup and the colour of the sample was determined by measuring L*, a* and b* values.

Plate 3.1 Hunter Lab colourimeter

Plate 3.2 pH meter

Plate 3.3 Water activity meter

3.2.2.5 Wettability

The wettability of the wall materials were measured by the procedure explained by Bae and Lee (2008) and Fuchs *et al.* (2006). One gram of each wall materials were taken and sprinkled over the surface 100 ml of distilled water at 20°C without agitation. The time taken for the powder particles to sediment or sink or submerse below and disappear from the surface of water were recorded and used for comparing the extent of wettability between different samples.

3.2.2.6 Cold Water Solubility

The cold water solubility of the wall materials was determined by the method described by Loksuwan (2007). One gram of wall materials were taken and mixed with 100 ml of water at room temperature for 30 minutes using a magnetic stirrer. A 10 ml aliquot of the supernatant solution was taken and transferred to a 15 ml centrifuge tube and then centrifuged for 15 minutes. The aliquot of the supernatant was then taken in a pre-weighed petridish, evaporated on a steam bath and dried in an oven at 110°C overnight. The cold water solubility was determined by using the following formula:

$$\text{Cold water solubility}(\%) = \frac{10 \times \text{Grams of solid in supernatent}}{\text{Grams of sample}} \times 100$$

.....3.3

3.3 EXPERIMENTAL DESIGN FOR THE OPTIMISATION OF THE WALL MATERIAL, CORE CONCENTRATION AND SPRAY DRYING TEMPERATURE FOR MICROENCAPSULATION AND NANOENCAPSULATION OF CUMIN OIL

After the selection of suitable wall materials and spray dryer inlet temperature from the preliminary studies for microencapsulation and nanoencapsulation, the independent and dependent variables were selected for the encapsulation.

In the present study, Response Surface Methodology (RSM) was employed in the experimental design. It is a collection of statistical and mathematical techniques which are useful for developing, improving and optimising processes. RSM is most extensively applied in the particular situations where a number of input variables potentially influence some performance measure or quality characteristic of the process. By careful

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design of experiments, the objective is to optimise an output variable or response which is influenced by a number of input variables or independent variables. An experiment is a series of tests, called runs, in which changes are made in the independent variables in order to identify the reasons for changes in the output response (Morshedi and Akbarian, 2014)

The experiments were designed based on Box–Behnken design in RSM with three factors at three levels (−1, 0 and +1). The independent variables or the process parameters selected for the optimisation were carrier blend ratio, core concentration or flavour load (percent w/w), and spray dryer inlet air temperature (°C) as explained above. The number of experiments (N) or runs in the Box–Behnken design is obtained from the equation $N = 2\ k\ (k - 1) + C_o$ (where k is number of factors and C_o is the number of central points). In the present study there were 17 experiments with 5 central points. The Design-Expert software (version 7.0.0) was employed for the statistical analysis.

3.3.1 Independent Variables:

(1) Carrier blend ratio (w/w)

Gum arabic : Maltodextrin

(a) C1: 1:2

(b) C2: 1:3

(c) C3: 1:4

(2) Core concentration (percent w/w)

(a) D1: 10 %

(b) D2: 20 %

(c) D3: 30 %

(3) Spray dryer inlet air temperature (°C)

(a) T1: 150

(b) T2: 160

(c) T3: 170

3.3.2 Dependent variables:

Product characteristics

(a) Moisture content

(b) Water activity

(c) Bulk density

(d) Powder wettability

(e) Cold water solubility

(f) Colour

(g) Encapsulation efficiency

3.3.3 Characteristics of Optimally Produced Encapsulated Cumin Oil Powders

(a) Particle size

(b) Active component retention

(c) Morphological characteristics

Table 3.1 Coded and un-coded values of independent variables in Box-Behnken design for microencapsulation and nanoencapsulation of cumin oil

Independent variables	Code variables	Levels in coded form		
		-1	0	+1
Carrier blend ratio	A	1:2	1:3	1:4
Core conc. (percent w/w) (%)	B	10	20	30
Inlet air temperature (°C)	C	150	160	170

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Table 3.2 Box-Behnken experimental design for the production of microencapsulated cumin oil powder

	Coded variables			Un-coded variables		
Run	**Carrier blend ratio**	**Core conc. (%)**	**Temperature (°C)**	**Carrier blend ratio**	**Core conc. (%)**	**Temperature (°C)**
1	0	0	0	3	20	160
2	+1	+1	0	4	30	160
3	0	+1	-1	3	30	150
4	0	0	0	3	20	160
5	+1	0	+1	4	20	170
6	0	0	0	3	20	160
7	-1	-1	0	2	10	160
8	-1	+1	0	2	30	160
9	-1	0	-1	2	20	150
10	-1	0	+1	2	20	170
11	0	0	0	3	20	160
12	0	-1	+1	3	10	170
13	0	0	0	3	20	160
14	0	-1	-1	3	10	150
15	0	+1	+1	3	30	170
16	+1	-1	0	4	10	160
17	+1	0	-1	4	20	150

Table 3.3 Box-Behnken experimental design for the production of nanoencapsulated cumin oil powder

	Coded variables			Temperature (°C Un-coded variables		
Run	**Carrier blend ratio**	**Core conc. (%)**	**)**	**Carrier blend ratio**	**Core conc. (%)**	**Temperature (°C)**
1	+1	+1	0	4.00	30.00	160.00
2	0	+1	-1	3.00	30.00	150.00
3	-1	0	-1	2.00	20.00	150.00
4	0	0	0	3.00	20.00	160.00
5	+1	0	+1	4.00	20.00	170.00
6	0	0	0	3.00	20.00	160.00
7	-1	-1	0	2.00	10.00	160.00
8	-1	+1	0	2.00	30.00	160.00
9	0	-1	-1	3.00	10.00	150.00
10	0	0	0	3.00	20.00	160.00
11	0	+1	+1	3.00	30.00	170.00
12	0	0	0	3.00	20.00	160.00
13	-1	0	+1	2.00	20.00	170.00
14	+1	-1	0	4.00	10.00	160.00
15	0	-1	+1	3.00	10.00	170.00
16	0	0	0	3.00	20.00	160.00
17	+1	0	-1	4.00	20.00	150.00

3.4 ENCAPSULATION OF CUMIN OIL BY SPRAY DRYING

Microencapsulation and nanoencapsulation of cumin oil was carried out using spray drying technique. In a spray dryer the active substance or the core material to be protected or encapsulated is initially homogenized with the coating agents or wall material at a given ratio in a liquid (water) phase. The emulsion thus formed is then atomised into the drying chamber of the spray dryer where the water gets evaporated by hot air and the coating or wall material, basically film-forming natural and synthetic biopolymers, covers or entraps microdroplets or microparticles of core compound. By the removal of water from the emulsion, dry capsules as free-flowing powders are formed and collected at the bottom of the spray dryer or in the powder collector of the cyclone. The main purpose of spraying is to increase the surface area of the material being sprayed, which thereby improves heat exchange between the solution and the drying agent (Janiszewska and Witrowa-Rajchert, 2009). The process flow chart for the production of both microencapsulated and naoencapsulated cumin oil powder is shown in Fig 3.1.

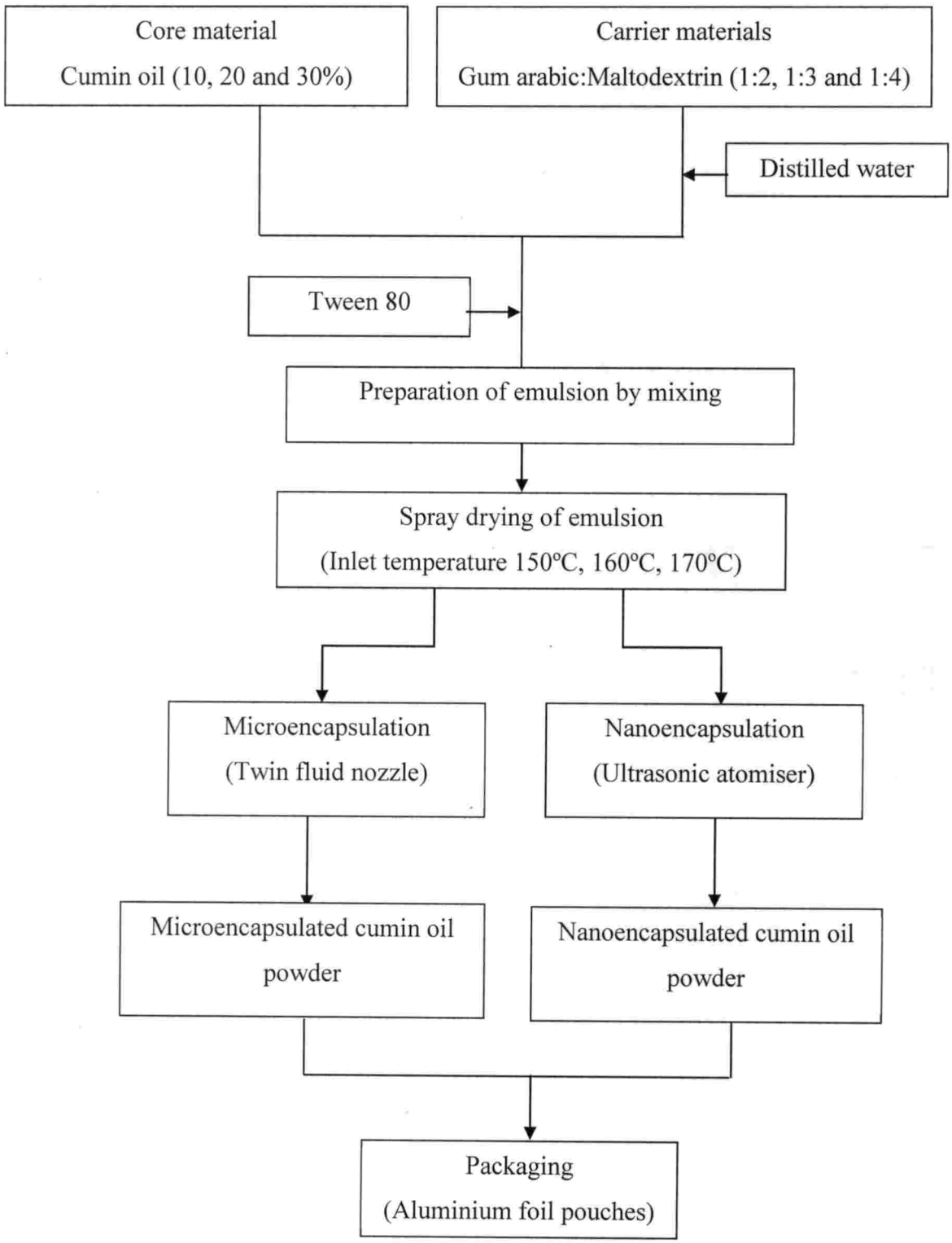


Fig. 3.1 Process flow chart for the production of spray dried encapsulated cumin oil powder

3.4.1 Microencapsulation of Cumin Oil

In the present research work, microencapsulation of cumin oil was carried out using a tall type spray dryer with twin fluid nozzle (M/s S.M. Scientech, Kolkata) (Plate 3.4) which is a co-current type having an evaporation rate of 1000 ml/h. The various parts of the spray dryer are air supply system, feed supply system, atomiser, drying chamber, powder recovery system and control panel.

3.4.1.1 Hot Air Supply System

Air supply system consists of compressor, air filter and air heater. The air is compressed by a compressor and this compressed air is introduced into twin fluid pressure nozzle atomiser after passing through an air filter and heater. The compressed air disintegrates the feed emulsion into a fine mist. An air filter is essential to cease the entry of microorganism. The air is heated through electric heating coils to get a maximum temperature up to 350°C.

3.4.1.2 Feed Supply System

The feed supply system consists of a peristaltic pump and a feed source. A beaker filled with the sample is the feed source. The peristaltic pump pumps the feed into the atomiser placed at the top of the spray dryer. The peristaltic pump consist of five rollers, which will squeeze the hypalon natural rubber tube (6 mm diameter) passing through them with its contents against the walls of the pump, which will create a vacuum behind each roller. This vacuum will suck the feed solution from the feed source. The motorised peristaltic pump has variable speed arrangement to control the flow. The rpm of the DC motor was controlled by a rotary knob.

3.4.1.3 Drying Chamber

The spray dryer consists of a vertical drying chamber which is made up of SS304 stainless steel. The drying chamber is cylindrical in shape with a conical bottom portion having gradual tapering for easy flow of dried powders. The inner side of the drying chamber is polished to 180-200 grit fineness to eliminate the accumulation of powders during operation. The atomiser is placed at the top most portion of the drying chamber. At the bottom of the conical portion a glass bottle of 1000 ml is flanged through teflon gaskets for collecting the dried powder particles.

3.4.1.4 Atomiser

The twin fluid nozzle placed at the top of the drying chamber will distribute the feed solution into the drying chamber in the form of fine spray from the ceiling of main chamber in downward direction. Along with the feed, compressed air will also be introduced into the twin fluid nozzle and the kinetic energy of this compressed air is utilized for dispersing the feed solution in to fine mist. Twin fluid nozzle atomisers can produce a wide range of flow rate and droplet size. A cone shaped spray pattern is produced inside the drying chamber by this nozzle. The compressed air pressure for the flow of the spray in the present study was kept at 3 kg/cm^2.

3.4.1.5 Powder Recovery System

Fine powder particles produced after the evaporation of moisture from the feed will be carried along with the hot air from the drying chamber into the cyclone separator where they will get separated. Air along with particles swirl in a spiral direction down the cyclone and, due to density difference, air leaves the cyclone through a duct pipe since it is less/denser than the particles and the particles get collected in glass bottle attached at the bottom of the cyclone separator through threaded flange with a teflon gasket.

3.4.1.6 Control Panel

The blower speed, feed rate and inlet and outlet temperature were controlled through an electrical control panel with appropriate regulators, ON/OFF push buttons and indicators. In addition to this an automatic and manual de-blocking knob is also connected to solve the clogging of atomiser.

There are two stages involved in the encapsulation process by the spray drying technique. The first stage is the formation of a stable emulsion of core material in a solution of carrier material with which it is immiscible with the addition of an emulsifier. The second stage is the atomisation of the prepared emulsion into drying chamber of the spray dryer and evaporation of water.

3.5 PREPARATION OF EMULSION

Two hundred and fifty grams of coating or wall materials (gum arabic and maltodextrin) were taken in the required proportions and are dissolved in 300 ml of distilled water by continuous stirring. After complete dispersion of the coating materials in the distilled water the final volume was made up to 500 ml by the addition of distilled water. The resultant 50% solid wall material solutions were filtered with muslin cloth in order to eliminate the foreign materials coming into the solution. The resultant solutions were then fortified with 25, 50 and 75 g of cumin oil to obtain a flavour load (core concentration) of 10, 20 and 30% (w/w) of the wall solids respectively (Fernandes *et al.*, 2008b). Two drops of Tween-80 (Polysorbate 80 or Polyoxyethylene (80) sorbitan monooleate) was added to enhance the emulsifying and film forming properties (Krishnan *et al.*, 2005). The mixture was then emulsified with a high-speed mixer until the cumin oil gets completely dispersed.

3.6 SPRAY DRYING OF THE PREPARED EMULSION

After preparing the feed solution, the spray dryer unit was heated to the desired unit. When the inlet air temperature get stabilised, the outlet temperature was adjusted and stabilised with distilled water which was achieved by varying the speed of feed pump. The feed flow rate of peristaltic pump was adjusted to 6 rpm and the compressed air pressure was adjusted to 3 kg/cm^2. The prepared emulsion was then pumped into the twin fluid nozzle and then sprayed to the drying chamber of the spray dryer. The spray dryer inlet air temperature used was varied from 150°C to 170°C as per the experimental design. The blower speed was adjusted to 1500 rpm. After the removal of moisture from the sprayed emulsion droplets, the encapsulated cumin oil powder was collected from the glass bottles of both drying chamber and cyclone separator which then mixed thoroughly and packed in aluminium foil pouches, sealed air tight using a hand sealer and stored at room temperature for further analysis.

3.7 NANOENCAPSULATION OF CUMIN OIL

Nanoencapsulation of cumin oil was carried out at the Nanotechnology laboratory, Department of Processing and Food Engineering, College of Agricultural Engineering, Raichur. The procedure adopted for nanoencapsulation of cumin oil was same as that of the microencapsulation. Twelve per cent carrier solids (wet basis) emulsions were prepared for nanoencapsulation of cumin oil. The spray drying was carried out in the laboratory spray dryer (Labultima LU-222 advanced, Labultima, Mumbai) (Plate 3.5) with ultrasonic atomiser. This spray dryer is a vertical co-current type with an evaporation rate of 1000 ml/h. The components of spray dryer include air filter, air heater and air distributor, feed supply system, atomiser, drying chamber, powder recovery system (consists of twin cyclone), scrubber, aspirator assembly and air compressor. Drying chamber has a diameter of 150 mm. The dimension of complete unit

is 40 x 40 x 50 cm (below lab table). Aspirator flow rate was kept at 40Nm^3/hr. The compressor was kept at 8 lb/$inch^2$ and the pressure gauge reading was kept at 2.5-3 bar.

For the generation of nanoparticles, a Programmable Ultrasonic Atomiser Drive (PUAD) with ultrasonic spray nozzle was used (Labultima, Mumbai). The ultrasonic spray nozzle consists of titanium vibratory cylinder. The minimum power was kept at 7.1 watts. A flow rate of 2 ml/min was kept constant for whole treatments.

Plate 3.4 Spray dryer used for the production of microencapsulated cumin oil powder

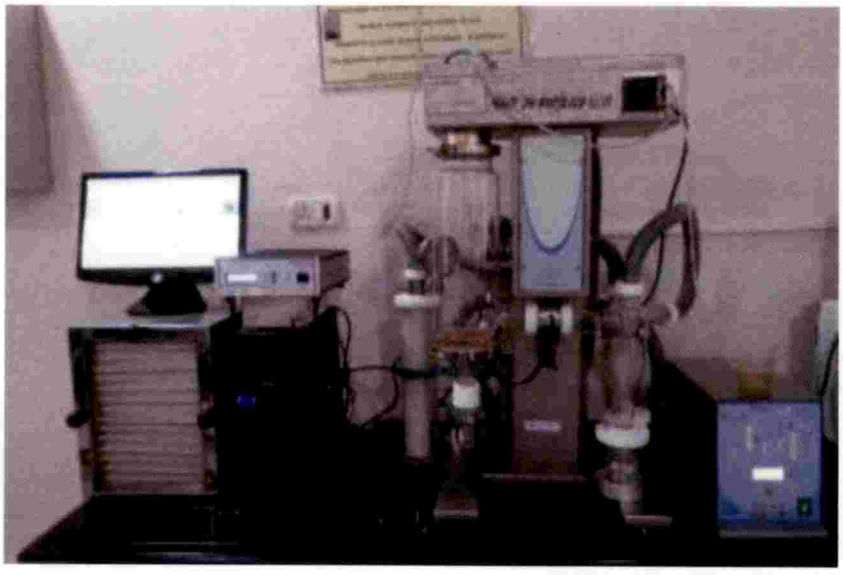


Plate 3.5 Spray dryer used for the production of nanoencapsulated cumin oil powder

3.8 QUALITY ANALYSIS OF MICROENCAPSULATED AND NANOENCAPSUALTED CUMIN OIL POWDERS

3.8.1 Moisture Content

Moisture content of the encapsulated powders was determined by the method as described in the section 3.2.2.1. The initial and final values of weights after drying were noted and used to calculate the moisture content of the encapsulated powders.

3.8.2 Water Activity

The water activity of encapsulated cumin oil powders were measured using Aqua lab water activity meter (model: Aqua lab, Decagon Devices Inc., Pullman (Wa), USA) (Plate 3.3) as explained I section 3.2.2.2.

3.8.3 Bulk Density

The bulk density of the both the encapsulated powders were found by the method as described in the section 3.2.2.3. The bulk density was expressed in g/cm^3.

3.8.4 Wettability

The wettability of the encapsulated cumin oil powders were found out by the method suggested by Bae and Lee (2008) and Fuchs et al. (2006) as explained in section 3.2.2.5. The time required for I g of the powder particles to sediment or sink or submerse below and disappear from the surface of 100 ml of distilled water were measured.

3.8.5 Cold Water Solubility

The cold water solubility of the encapsulated cumin oil powders were calculated by the method explained by Loksuwan (2007) as explained in section 3.2.2.6.

3.8.6 Colour

The colour of the encapsulated cumin oil powders were found out using Hunter lab colour flex meter (Hunter Association laboratory, Inc., Reston, Virgina, USA) (Plate 3.1).

3.8.7 Encapsulation Efficiency

Encapsulation efficiency is considered as one of the most important quality characteristics in encapsulation of oils. It is defined as the percentage of encapsulated oil in total oil. Encapsulation efficiency (EE) was calculated by determining the amount of total oil and surface oil content in the encapsulated powders using the methods described below:

3.8.7.1 Total Oil

A Clevenger hydrodistillation apparatus was used for determining the amout of total oil present in the encapsulated cumin oil powders. Ten grams of the encapsulated cumin oil powder was accurately weighed and dissolved in 150 ml of distilled water in a 250 ml flask. The flask was manually shaken for one minute to break the clumps and facilitate dissolution of the powder. A Clevenger oil trap and a water cool condenser were attached. Then the solution was allowed to boil and distil for 3 h. The volume of the oil was read directly from the oil collection arm and then multiplied by its specific gravity for converting to weight.

Plate 3.6 Clevenger hydrodistillation apparatus

3.8.7.2 Surface Oil

The surface oil or unencapsulated oil or free oil content was determined by the method explained by Bea and Lee (2008) and Tan *et al.* (2005) with some modifications. Fifty millilitres of hexane was added to 10 g of accurately weighed encapsulated cumin oil powder in a 100 ml flask with a screw cap and shaken for 2 minutes at ambient temperature for extracting the unencapsulated oil. After that the mixture was filtered through a Whatman No.1 filter paper. The collected washed encapsulated powder on the filter paper was again three times rinsed with 20 ml of hexane at each

time by passing it through the powder. The residual powder was then dried to vapourise all residual solvent or hexane at 60°C until a constant weight was achieved. The surface oil content was then calculated as percentage by the weight difference in the powder before and after extraction and washing with hexane. Encapsulation Efficiency (EE) was calculated using the following formula:

$$\text{Encapsulation efficiency}(\%) = \frac{\text{Total oil} - \text{Amount of surface oil}}{\text{Total oil}} \times 100$$

.....(3.4)

3.9 CHARACTERISATION OF OPTIMALLY PRODUCED ENCAPSULATED POWDERS

The optimisation of process parameters on the basis of the quality analysis of both microencapsulated and nanoencapsulated cumin oil powders were done and the optimally produced powder sample was analysed for the following:

3.9.1 Particle Size Analysis

The particle size analysis of the optimally produced microencapsulated and nanoencapsulated powders were carried out at 'Center for nanotechnology' laboratory, UAS, Raichur. The Zetasizer nano range (ZETA Sizer, nano383 issue 5.0, Malvern, England) (Plate 3.7) was used for the particle size measurement. These instruments provide the ability to measure three characteristics of particles or molecules in a liquid medium. These three fundamental parameters are *viz.*, particle size, zeta potential and molecular weight. By the use of Zetasizer system, these three parameters can be measured over a wide range of concentrations. Zetasizer (nano series) was used in the study as dynamic light scattering apparatus. The range of particle size that can be measured with this instrument is 0.6 nm – 6 μm. The sample needs to be diluted for using the equipment.

Computer system uses the Malvern software for controlling of the equipment and for analysing the results.

In the present study the particle size measurements were performed on aqueous suspensions of optimally produced both microencapsulated and nanoencapsulated cumin oil powders. The procedure for the preparation of aqueous suspensions was as follows: one gram of both encapsulated powder samples were carefully weighed and then dispersed in 10 ml of distilled water. The suspension was stirred by the mechanical stirrer for at least 10 minutes for breaking powder agglomerates which results in fine, colloidal particles completely dispersed in water. A small portion of the sample was taken into a disposable cuvette and placed in the Zetasizer for measurements. Care was taken for avoiding the air bubbles in the sample while filling the cuvette.

3.9.2 Morphological Characteristics

The Scanning Electron Microscopy (SEM) analysis was done at 'Center for nanotechnology' laboratory, UAS, Raichur with a scanning electron microscope (Carl Zeiss, Zeiss group, Oberkochen, Germany) (Plate 3.8). SEM analysis was used to examine the morphological characteristics of optimised samples of both microencapsulated and nanoencapsulated cumin oil powders according to the modified methodology described by Jafari *et al.* (2008). Small amounts of encapsulated powders were taken from well mixed powder samples and these powders were attached to the surface of a double-sided adhesive carbon tape fixed to SEM stubs. The samples were subsequently coated with a thin layer of gold-palladium under vacuum with the help of a sputter coater (Plate 3.9). Plasma current of 10 mA for a period of 90 s were used for the coating of the samples. A Zeiss scanning electron microscope (EVO LS 10, Carl Zeiss company, Germany) was used for analyzing the morphology of samples which were systematically observed at 2000X and 6000X magnification.

3.9.3 Active Component Retention

The main active components of Indian cumin oil are cuminaldehyde and γ –terpinene (Dubey *et al.*, 2017). From the optimally produced microencapsulated and naoencapsulated cumin oil powders, the main active components were determined by gas chromatography (Shimadzu GC-17A, Japan) (Plate 3.10). Cuminaldehyde and γ –terpinene standards were procured from M/s.Sigma, St. Louis, MO, USA. For finding out these compounds the procedure described by Soottitantawat *et al.* (2003) and Liu *et al.* (2001) with slight modification. Two grams of the encapsulated cumin oil powders were dispersed in 10 ml of water taken in a glass bottle to which 2 ml of hexane was then added. After this the mixture is violently mixed by using a vortex mixer for about one minute. After mixing properly the mixture was heated in a heating block with intermittent shaking for extracting the flavour into the organic solvent. The extraction time and temperature were 30 min and 60°C respectively. By completing the extraction process the mixture was then centrifuged at 3000 rev/min for 10 min in order to separate the organic phase from the water. Two microliters of the organic phase were injected for each sample into the gas chromatograph. The chromatographic conditions used were as follows: flame ionization detector (FID) at 240°C with N_2 as the carrier gas, an oven temperature programme comprising of an injector temperature of 230°C, initial temperature of 55°C for 0 min, temperature increase was from 55 to 90°C at 6°C/min, held for 1min, then 90 to 118°C at 6°C/min, 118–120°C at 0.3°C/min, 120 to 230°C at 10°C/min (Kanakdande *et al.*, 2007)

Plate 3.7 Zetasizer

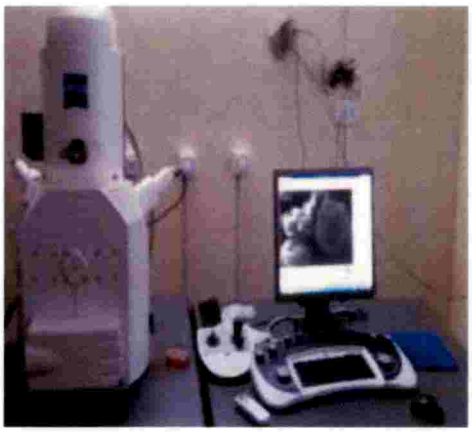


Plate 3.8 Scanning electron microscope

Plate 3.9 Sputter coater

Plate 3.10 Gas Chromatography

3.10 COST ANALYSIS FOR THE PRODUCTION OF MICROENCAPSULATED AND NANOENCAPSULATED CUMIN OIL POWDERS

The cost analysis for the production of both microencapsulated and nanoencapsulated cumin oil powders by the spray drying technique was estimated by considering the fixed, variable and other related costs which include the following costs *viz.,* cost of building, spray dryer, raw materials, processing, labour, electricity and other related costs. The cost of operation was determined by estimating the fixed cost and variable cost. The variable cost of unit was calculated by considering electricity charges, repairs and maintenance, cost of labour and raw materials.

Results and Discussion

CHAPTER IV

RESULTS AND DISCUSSION

This chapter deals with the results and discussion of the experiments conducted on microencapsulated and nanoencapsulated cumin oil powders by spray drying technique. The effect of various process parameters on the dependent variables are discussed in detail.

4.1. PHYSICO-CHEMICAL PROPERTIES OF RAW MATERIALS

4.1.1 Physico-chemical properties of cumin oil

The various physico-chemical properties of cumin oil procured from Vaidyaratnam P.S. Varier's Arya Vaidya Sala, Kottakkal, Kerala, India, were determined by the standard procedures. Data thus generated were statistically analysed and tabulated in Table 4.1. The pH value of cumin oil was 3.3 and specific gravity was 0.90. The colour values L*, a* and b* were 1.8, 0.29 and 0.81 respectively.

Table 4.1 Physico-chemical properties of cumin oil

Physico-chemical properties		**Value**
pH		3.30±0.007
Colour	L*	1.8±0.142
	a*	0.29±0.134
	b*	0.81±0.212
Specific gravity (at 25°C)		0.90±0.007

4.1.2 Physico-chemical Properties of Wall Materials

The physico-chemical properties of gum arabic and maltodextrin, as carrier agents, were found out as per the procedure explained in 3.1.2 and average values are tabulated in Table 4.2. It was observed that the gum arabic is more hygroscopic in nature which is having moisture content of 4.10 whereas in the case of maltodextrin it was only 3.84%. The water activity values of gum arabic and maltodextrin were recorded as 0.365 and 0.341 respectively. The bulk density values were found out as 0.615g/cm^3 for gum arabic and 0.322 g/cm^3 for maltodextrin. The L*, a*, b* values of gum arabic were 83.36, 2.38 and 11.21 and that of maltodextrin were 95.65, -0.50 and 2.58 respectively. The reconstitutional properties like wettability and solubility were higher in the case of gum arabic than maltodextrin.

Table 4.2 Physico-chemical properties of wall materials

Physico-chemical properties		**Gum arabic**	**Maltodextrin**
Moisture content (% w.b.)		4.10±0.011	3.84±0.061
Water activity		0.365±0.009	0.341±0.006
Bulk density (g/cm^3)		0.615±0.037	0.322±0.011
Colour	L*	83.36±0.558	95.65±0.809
	a*	2.38±0.009	-0.51±0.013
	b*	11.21±0.036	2.58±0.010
Wettability (s)		115.94±8.15	130.5±7.20
Solubility (%)		97.09±0.76	94.92±1.64

These properties directly or indirectly affect the quality of the final product.

4.2 OPTIMISATION OF THE WALL MATERIAL, CORE CONCENTRATION AND SPRAY DRYING TEMPERATURE FOR MICROENCAPSULATION AND NANOENCAPSULATION OF CUMIN OIL

Box-Behnken experimental design was used for the selection of best combination of process parameters in the present encapsulation processes. Microencapsulation and nanoencapsulation of cumin oil was carried out by the spray drying method with different combinations of gum arabic and maltodextrin (carrier blend ratio), core concentration and spray dryer inlet temperature as described in the section 3.3 and the encapsulated powder obtained are shown in Plate 4.1. These processes were optimised by investigating the effect of process parameters (carrier blend ratio, core concentration and spray dryer inlet temperatures) on the quality of the final product. Seventeen experiments were carried out for both microencapsulation and nanoencapsulation. The quality of the microencapsulated and nanoencapsulated powders were assessed in terms of moisture content, water activity, bulk density, wettability, cold water solubility, colour and encapsulation efficiency. Analysis of variance (ANOVA) was conducted for determining the significant effects of independent variables or process variables on each dependent variables or responses. The significance of each coefficient was checked by using p-values. The p value was also used to understand the pattern of mutual interactions between the independent variables. The model terms become significant when the p value is less than 0.05. The smaller the magnitude of the 'p', the more significant is the corresponding coefficient. The adequacy of regression model was checked by R^2, Adjusted R^2, Adequate Precision and Fisher's F-test (Montgomery, 2001).

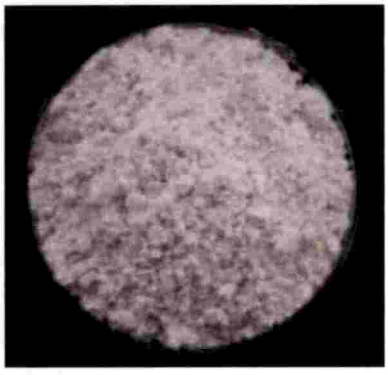
Adjusted R^2 is a measure of the amount of variation around the mean explained by the model, adjusted for the number of terms in the model. As the number of terms in the model increases, the adjusted R^2 decreases if

those additional terms do not add value to the model. Adequate precision compare the range of predicted values at design points to the average prediction error.

The significance of all terms in the polynomial was judged statistically by computing the F-value at probability (p) of 0.1 to 0.01. A complete second order quadratic model was employed to fit the data. R^2, Adjusted R^2, predicted R^2 (a measure of how good the model predicts a response value) and Fischer F-test were used to test the adequacy of the model (Haber and Runyon, 1977). If the value of R^2 becomes smaller, then there is less relevance the dependent variables in the model have in explaining the behaviour variation. Optimisation of process variables was done by partially differentiating the model with respect to each parameter, equating to zero and simultaneously solving the resulting function. The regression coefficients were then used to make statistical calculation to generate three-dimensional plots for the regression model.

Three-dimensional response surface was plotted to visualise the relationship between the significant ($p<0.05$) interaction effects of independent variables and response variables.

(a)



(b) **(c)**

Plate 4.1 (a) Cumin oil, (b) Microencapsulated cumin oil powder, (c) Nanoencapsulated cumin oil powder

4.2.1.1 Moisture Content

Moisture or water content indicates the total amount of water present in a food. It is one of the important factors which can affect the stability and caking property of the products during the period of storage and distribution. Moisture content and temperature will affect the shelf life of dried microcapsules (Quispe-Condori *et al.*, 2011). Dried foods with moisture content between 3 and 10% will have good stability during storage (Barroso *et al.*, 2014).

In the present work, the effect of three independent variables (core concentration, carrier blend ratio and spray dryer inlet temperature) on moisture content of both microencapsulated and nanoencapsulated cumin oil powders produced are tabulated in Appendix A1 and B1 respectively.

The moisture content of microencapsulated cumin oil powders varied from 3.68 to 5.55% (w.b.) whereas for nanoencapsulated powders, it was from 2.7 to 4.23 %. In both cases, the higher moisture content was observed for the powder produced with a gum arabic:maltodextrin ratio of 1:2, 20% core concentration and a spray dryer inlet temperature of 150°C. The minimum value was recorded for the powder produced with a gum arabic:maltodextrin ratio of 1:4, 20% core concentration and at 170°C spray dryer inlet temperature.

The effect of carrier blend ratio, core concentration and spray dryer inlet temperature on moisture content of microencapsulated and nanoencapsulated cumin oil powders are illustrated by plotting 3D graphs representing the response surface generated by the model (Equation. 4.1 and 4.2) and are shown in Fig.4.1 and 4.2.

The spray dryer inlet temperature greatly affected the moisture content of encapsulated cumin oil powders. It is clear from the Fig.4.1 and 4.2 that the moisture content showed a decreasing tendency when the spray dryer inlet temperature was increased from 150 to170°C. The reason may be the relative humidity of the air present inside the spray dryer. When the difference between inlet and exit air temperatures in the spray dryer decreases, the relative humidity of the dryer air decreases, thereby moisture content of the product decreases. Similar observation was also reported by Reineccius (2004) and Finney *et al.* (2002).

It may be observed from Appendix A (Table A2) and Appendix B (Table B2) that core concentration did not show any significant effect on the moisture content of the encapsulated powders in the present study. Dian *et al.* (1996) in microencapsulation of palm-based oil and Hogan *et al.* (2001) in microencapsulation of soya oil also reported the same trend.

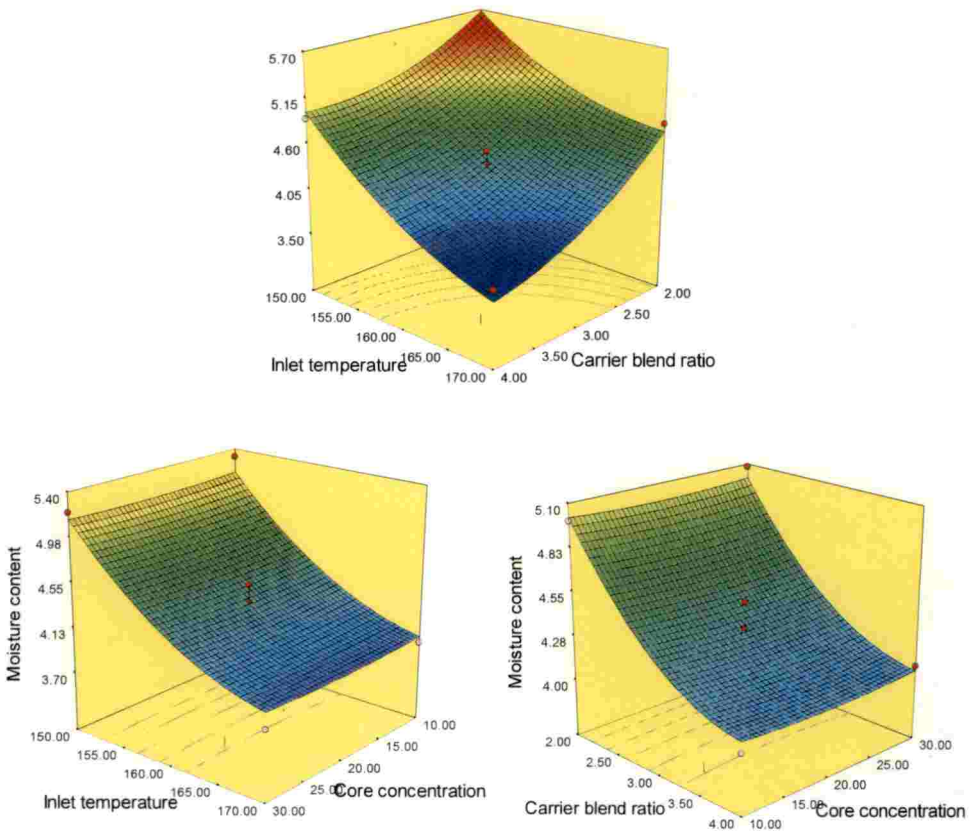


Fig. 4.1 Effect of process variables on moisture content of microencapsulated cumin oil powder

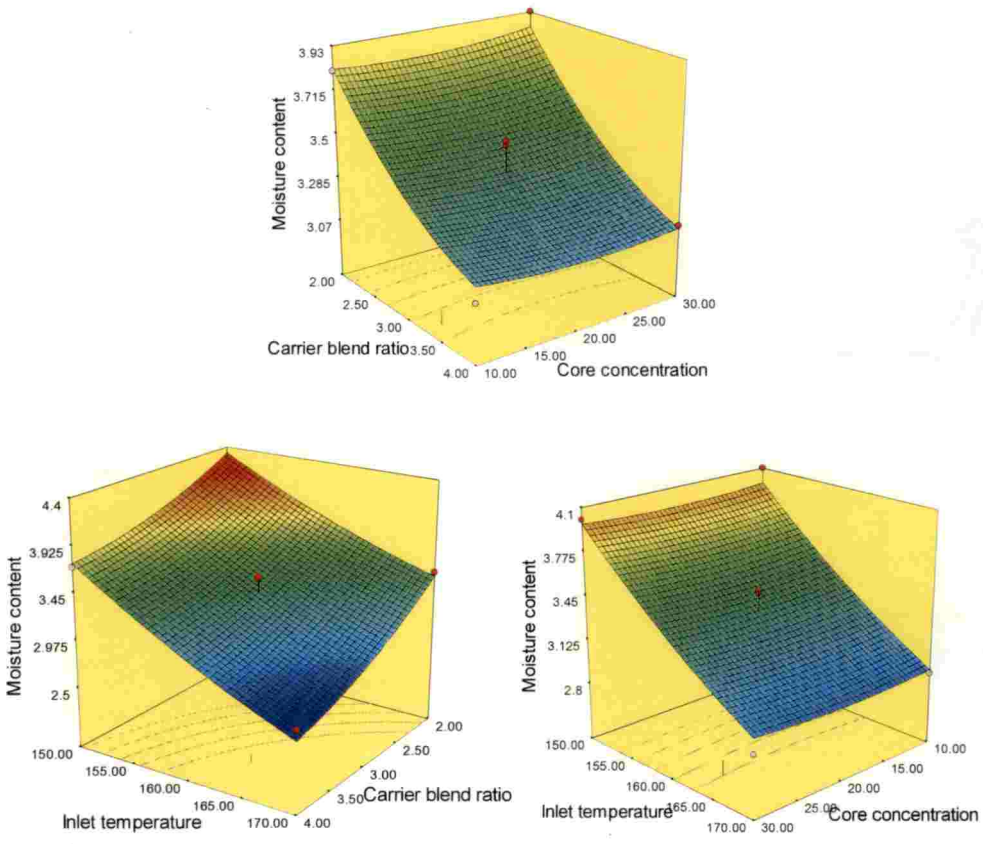


Fig. 4.2 Effect of process variables on moisture content of nanoencapsulated cumin oil powder

The type of wall material also affected the moisture content of both microencapsulated and nanoencapsulated cumin oil powders. It is evident from the Fig.4.1 and 4.2 that the moisture content of the encapsulated powders containing higher proportions of gum arabic was higher whereas the moisture contents of the product powder decreased when the proportion of maltodextrin in the emulsion increased. The reason for high moisture content occurred with increased gum arabic proportions may be due to its high hygroscopicity (Tonon *et al.*, 2012). Thus after the drying process, it absorbs water vapour directly. These results are consistent with the findings of Apintanapong, and Noomhorm, (2003) in the microencapsulation of 2-acetyl-1-pyrroline.

A second order non-linear regression equation was fitted between dependent and independent variables using the experimental values. Following regression models were obtained to predict the moisture content of microencapsulated cumin oil powder (4.1) and naoencapsulated cumin oil powder (4.2).

Moisture content =4.31-0.47A-2.500E-003 B-0.60C+0.000AB-0.12AC

$$+5.000E\text{-}003BC+0.21A^2+0.024B^2+0.22C^2 \quad \text{.....(4.1)}$$

Moisture content =3.36-0.35A+3.750E-003B-0.52C-0.013AB-0.060AC

$$+5.000E\text{-}003BC+0.096A^2+0.031B^2+0.069C^2 \quad \text{.....(4.2)}$$

Where A: Carrier blend ratio (gum arabic:maltodextrin)

B: Core concentration or flavour load (%) and

C: Spray dryer inlet temperature (°C)

The ANOVA table for moisture content of microencapsulated and nanoencapsulated cumin oil powders are shown in Appendix A (Table A2) and B (Table B2). For moisture content of the microencapsulated cumin oil powder the coefficient estimates and the corresponding p-values suggest that, among the test variables used in the study, the linear terms, A (gum arabic:maltodextrin), C (spray dryer inlet temperature) and quadratic terms A^2 and C^2 have significant effect. The p-value of the model was 0.0002 which implies that the model fitness was significant. The best fit model was expressed by the coefficient of determination R^2 (0.9674) obtained from Table A2, indicating that 96.74 per cent of the variability of the response could be explained by the model.

For moisture content of the nanoencapsulated cumin oil powders, the magnitude of p-value indicates that the linear terms, A and C have

significant effect. The p- value of the model was 0.0005 which implies that the model fitness was significant. In this case the R^2 value was 0.9576.

4.2.1.2 Water Activity

Water activity is a measure of the available water in a food system. It is the ratio of the vapour pressure in food and the saturated vapour pressure of water at the same temperature. It significantly influences the shelf life of powder particles. High water activity shortens the shelf life of the products due to high free water available for biochemical degradations. at water activity (a_w) lower than 0.6, the deterioration of dried powders caused by biochemical reactions and microorganisms can be prevented (Tang and Yang, 2004; Kha *et al.*, 2010).

The water activity of both microencapsulated and nanoencapsulated cumin oil powders obtained from various combinations of process parameters are tabulated in Appendix A (Table A3) and Appendix B (Table B3) respectively. The water activity of microencapsulated cumin oil powders varied between 0.261 and 0.385. The highest water activity was obtained for the powder produced with a gum arabic:maltodextrin ratio of 1:2, 20% core concentration and at 150°C spray dryer inlet temperature whereas the lowest value was obtained for those produced with 1:4 ratio of gum arabic:maltodextrin, 20% core concentration and at 170°C. In case of nanoencapsulated cumin oil powders the water activity varied between 0.205 and 0.338. The highest and lowest values of water activity were obtained for the same treatment combinations as in case of microencapsulation.

The values obtained for water activity of encapsulated powders with different combinations of process variables are presented in Fig.4.3 and 4.4 respectively. The results obtained showed that the effect of the different process variables on water activity of the encapsulated cumin oil powders showed the same effect as that of moisture content.

A second order non-linear regression equation described the relation between dependent and independent variables. The following regression models were obtained to predict the water activity of microencapsulated cumin oil powder (4.3) and nanoencapsulated cumin oil powder (4.4).

Water activity =0.30-0.032A+1.250E-004B-0.036C-0.010AB-1.000E

-002AC+8.000E-003BC+7.975E-003A^2+8.475E-003B^2+0.021C^2

.....(4.3)

Water activity = 0.24-0.032A+2.500E-004B-0.044C+4.250E-003AB

-3.000E-003AC+2.750E-003BC+0.014A^2+6.300E-003B^2+0.017C^2

.....(4.4)

ANOVA was performed to evaluate the significance of the coefficients of the quadratic polynomial models and it is given in Appendix A (Table A4) and B (Table B4) for microencapsulated and nanoencapsulated cumin oil powders respectively. The coefficient estimates and the corresponding p-values suggest that, the linear terms, A and C and the quadratic term C^2 were significant model terms. The p-value of the model was 0.0059, which indicated that the model fitness was significant. From Table A4, it can be concluded that the R^2 value is 0.9118.

For nanoencapsulated cumin oil powders, coefficient estimates and p-values suggest that the linear terms, A, C and C^2 were significant model terms. The p-value of the model was 0.0007, which indicated that the model fitness was significant. The R^2 value noted was 0.9532.

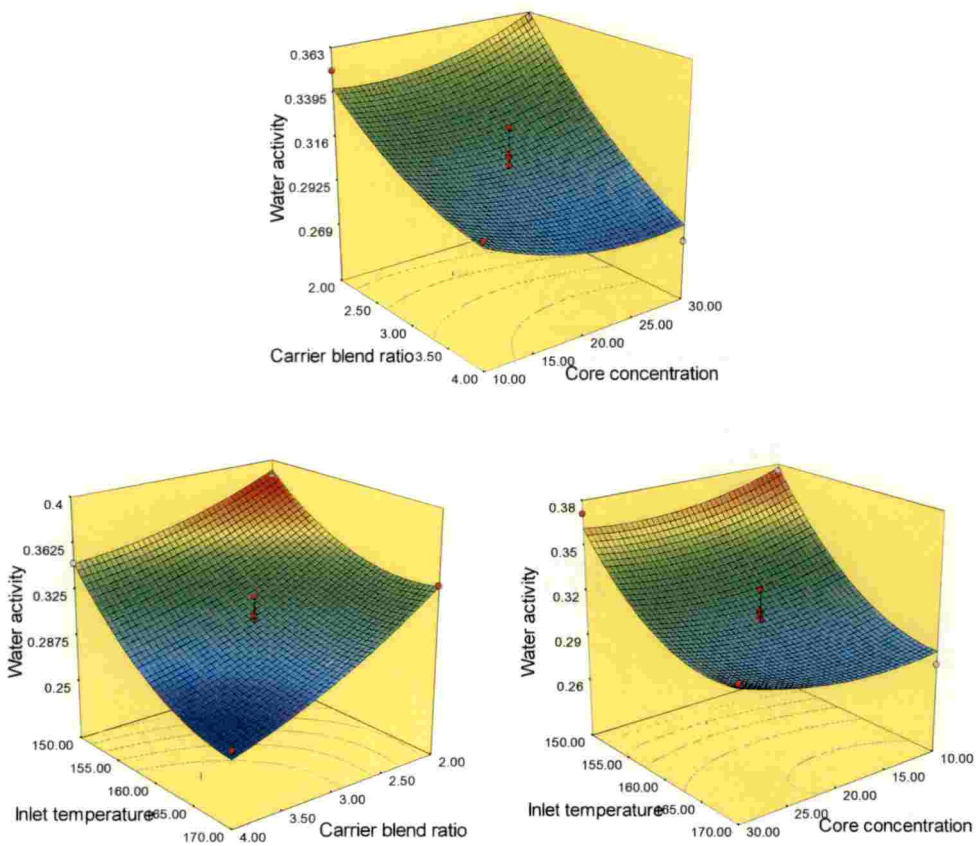


Fig. 4.3 Effect of process variables on water activity of microencapsulated cumin oil powder

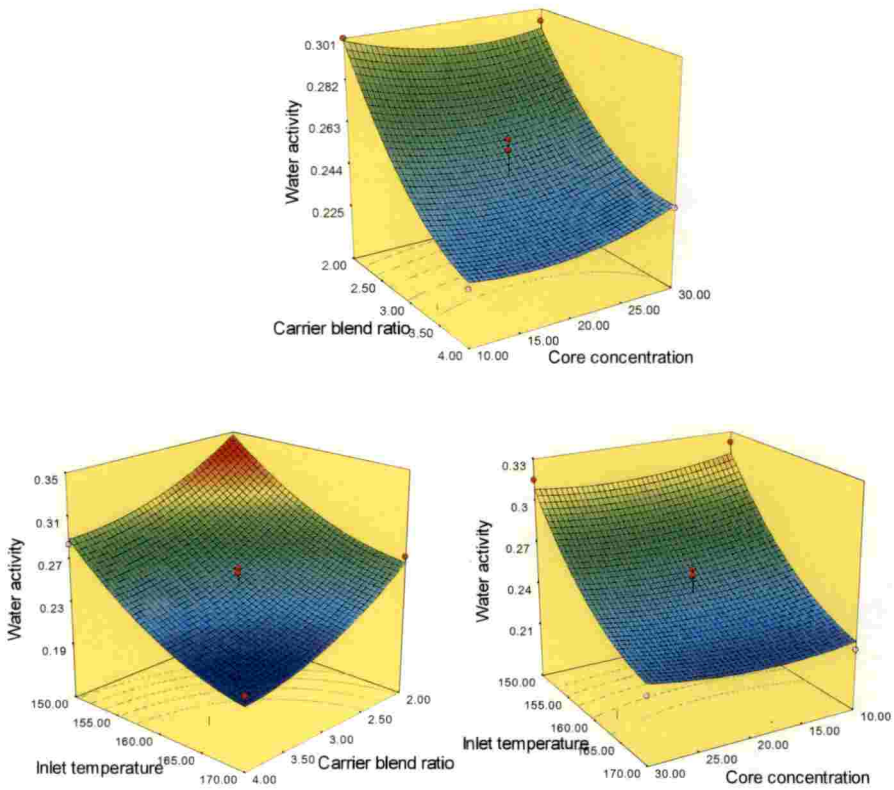


Fig. 4.4 Effect of process variables on water activity of nanoencapsulated cumin oil powder

4.2.1.3 Bulk Density

Bulk density is an important factor in packaging and shipping considerations because this parameter determines how much material, by weight, can fit into a container of specified volume (Finney *et al.*, 2002). As volume decreases, the density increases at a given constant mass. From a theoretical point of view, particles having smaller diameter contribute to a tighter packing and thereby results in an increased bulk density which indicates that the bulk density will be influenced by the particle size and geometry. Thus, spherical particles will pack the best and possess highest bulk densities, all other factors being equal. The powder having higher bulk density has the advantage that large amount of powder particles can be stored into smaller containers when compared to powder with lower values

of bulk densities. Higher bulk density may point out lower amount of air occluded in the spaces between particles (Carneiro *et al.*, 2013).

The effect of the three independent process variables *viz.*, core concentration, carrier blend ratio and spray dryer inlet temperature on the bulk density of both microencapsulated and nanoencapsulated cumin oil powders produced with different combinations of experiments are tabulated in Appendix A (Table A5) and B (Table B5) respectively.

The bulk density of microencapsulated cumin oil powder varied between 0.317 to 0.470 g/cm^3. In this case, the highest bulk density value was obtained for the powder produced with 1:2 gum arabic:maltodextrin ratio, 20% core concentration and a spray dryer inlet temperature of 150°C. The minimum value was recorded for the powder produced with a gum arabic:maltodextrin ratio of 1:4, 20% core concentration and at 170°C spray dryer inlet temperature.

For nanoencapsulation of cumin oil powder, the bulk density varied between 0.370 to 0.495 g/cm^3. The maximum value was obtained for the powder produced with gum arabic:maltodextrin ratio of 1:2, 10% core concentration and a spray dryer inlet temperature of 160°C. The minimum value was obtained for the powder produced with a gum arabic:maltodextrin ratio of 1:4, 20% core concentration and at 170°C spray dryer inlet temperature.

The effect of different process parameters on bulk density of microencapsulated and nanoencapsulated cumin oil powders are shown in 3D graphs representing the response surface generated by the model (Equation. 4.5 and 4.6) in Fig.4.5 and 4.6 respectively.

It was noted that the spray dryer inlet temperature significantly affected the bulk density of encapsulated cumin oil powders. An increase in the spray dryer inlet temperature reduced the bulk density of the

encapsulated cumin oil powders. This decrease in bulk density with the increase in spray dryer inlet temperature leads to the formation of increased particle size and it may be due to "skinning" over or casehardening of the emulsion droplets at higher temperatures. This may result in the formation of vapour-impermeable films on the drop surface. Consequently vapour bubbles are formed and finally droplet expansion takes place. These findings are consistent with the results of various works (Goula *et al.*, 2004; Jumah *et al.*, 2000; Chegini and Ghobadian, 2005).

The bulk density of both the encapsulated powders showed a direct relation to the proportion of gum arabic, ie., bulk density decreased with the decrease in the amount of gum arabic. The reason for this may be that increasing the gum arabic content will increase the moisture content of the encapsulated powders as stated earlier, and this will result in an increase in bulk density since the density of water is higher compared with the dry solid (Aghbashlo *et al.*, 2013). The same effect was also observed by Carneiro *et al.* (2013) for the microencapsulation of flaxseed oil. It may also be due to the fact that increased maltodextrin proportion might have increased the volume of air that get trapped in the particles since maltodextrin is skin forming and the powder particles produced by spray drying often contain air bubbles, due to the deposition of air present initially in the liquid feed or absorbed during the process of atomisation. In general, an increment in the trapped air volume will decrease the apparent density of the powder particles. This particle apparent density primarily determines bulk density of the powder (Krishnaiah *et al.*, 2014).

Bulk density was also affected by core concentration. When the concentration of oil in the emulsion was increased, the bulk density decreased. This is due to the fact that particle size is highly dependent on surface-free oil. Encapsulated powders containing higher amounts of surface or unencapsulated oil on the surface (therefore, a greater amount of extractable oil) tends to stick together and form lumps resulting in reduced

bulk density (Konstance *et al.*, 1995: Vega *et al.*, 2005). Domian and Wasak (2008) also observed the same in the microencapsulation of rapeseed oil.

The second order non-linear regression equation was fitted between dependent and independent variables using the experimental values. Following regression models were obtained to predict the bulk density of microencapsulated cumin oil powder (4.5) and nanoencapsulated cumin oil powder (4.6).

Bulk density = 0.35-0.026A-0.020B-0.037C-9.500E-003AB-0.021AC-

$$0.010BC+0.025A^2+0.052B^2+0.038C^2 \qquad \text{.....(4.5)}$$

Bulk density = +0.40-0.018A-0.025B-0.031C-4.500E-003AB-0.014AC

$$-1.000E-002BC+0.017A^2+0.036B^2+0.032C^2 \qquad \text{.....(4.6)}$$

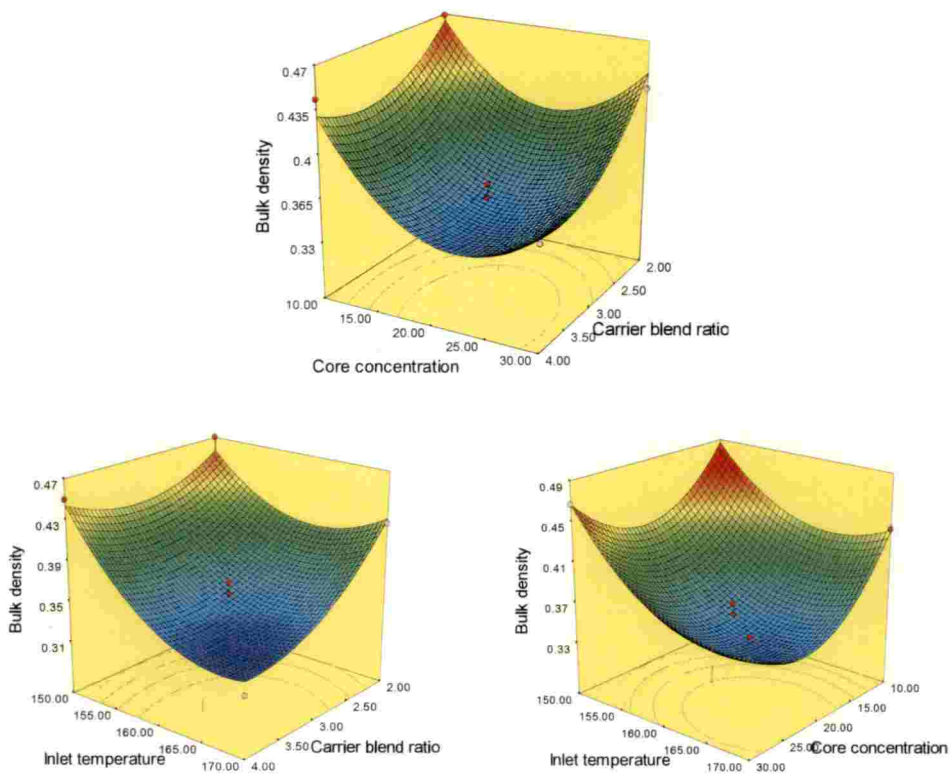


Fig. 4.5 Effect of process variables on bulk density of microencapsulated cumin oil powder

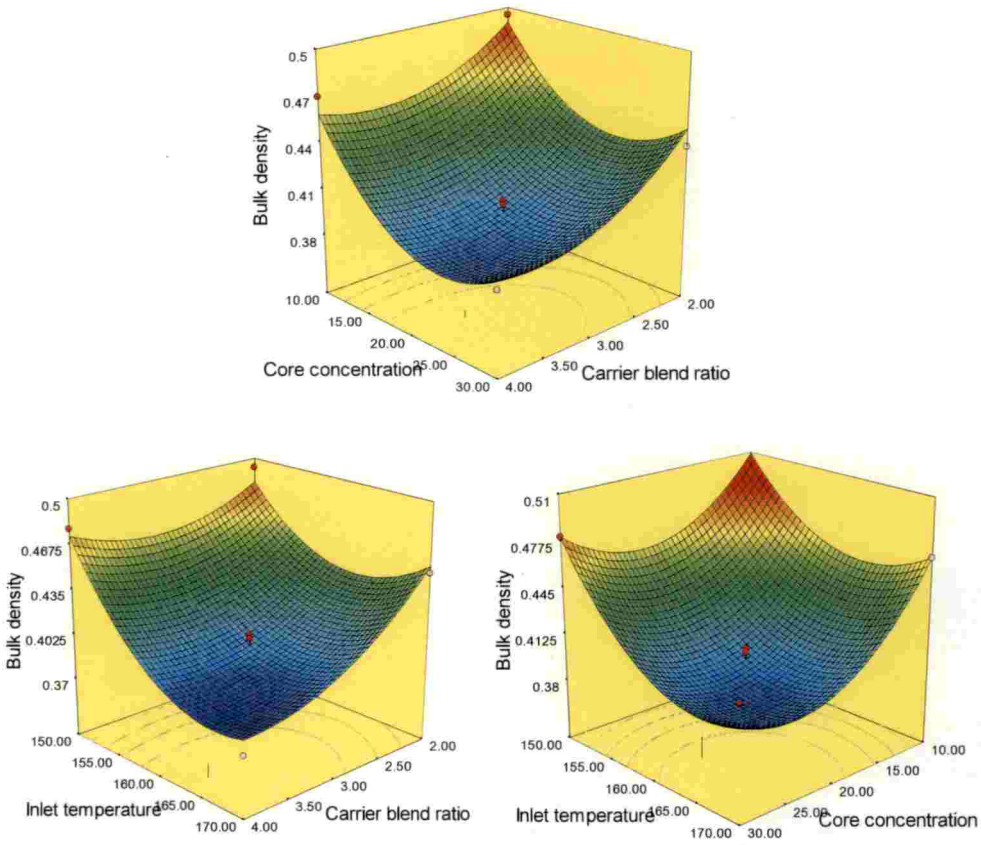


Fig. 4.6 Effect of process variables on bulk density of nanoencapsulated cumin oil powder

ANOVA performed to evaluate the significance of the coefficients of the quadratic polynomial models is given in Appendix A (Table A6) and B (Table B6) for microencapsulated and nanoencapsulated cumin oil powders respectively. For the bulk density of microencapsulated cumin oil powder, A, B, C, AC, A^2, B^2 and C^2 were significant model terms. The p-value of the model was 0.0004. From Table A6, the R^2 value was 96.11%.

For nanoencapsulated powder, A, B, C, A^2, B^2 and C^2 were significant model terms. The p-value of the model was 0.0008. From Table B8, it may be inferred that the value of R^2 was 95.24%.

4.2.1.4 Wettability

Reconstitution of the powder particles are one of the major properties of dehydrated emulsions. The dispersion of powder in liquids can be divided into four steps: wetting, submersing, dispersing and dissolving (if soluble) (Freudig *et al.*, 1999). Wettability of the powder particles is termed as the ability of bulk powder to imbibe a liquid under the influence of capillary forces. As the liquid penetrates the pores between the powder particles, the capillary pressure is opposed by the pressure drop in the flow direction. The parameters influencing the wettability of the products are powder particle size, porosity, density, surface area, surface charge, the presence of amphipathic substances, and the surface activity of the particles. In the present research work, the degree of wettability of the powder was measured by the time taken for powders to disappear from the water surface.

The wettability of microencapsulated and nanoencapsulated cumin oil powders produced with different treatments are tabulated in Appendix A (Table A7) and B (Table B7) respectively.

The wettability of microencapsulated cumin oil powder varied between 120.3 to 275 s. The minimum value of wettability was obtained for the powder produced with a gum arabic:maltodextrin ratio of 1:2, 20% core concentration and a spray dryer inlet temperature of 150°C. The maximum value was recorded for the powder produced with a gum arabic:maltodextrin ratio of 1:4, 20% core concentration and at 170°C spray dryer inlet temperature. In case of nanoencapsulation of cumin oil the values of wettability varied between 150.2 and 364.8 s. The minimum value of wettability was obtained for the powder produced with 1:2 ratio of carrier blend, 10% core concentration and at 160°C spray dryer inlet temperature whereas the maximum value was obtained from the treatment with 1:4 ratio of carrier blends, 20% core concentration and at 170°C spray dryer inlet temperature.

The effects of the process variables on wettability of the encapsulated powders are illustrated in 3D graphs representing the response surface generated by the model (Equation. 4.7 and 4.8) and are shown in Fig.4.7 and 4.8.

An increasing in the inlet air temperature and maltodextrin proportion resulted in an increase in average time to wet (i.e., the decrease in wettability). The reason for this increase in time may be the reduced residual moisture content in the product (Chegini and Ghobadian, 2005; Fernandes *et al.*, 2013). Caking, usually occurs in powders containing higher amount of moisture and these moisture can contribute to the wettability since the liquid penetrates into the pores more easily (Buffo *et al.*, 2002; Fernandes *et al.*, 2013).

Increasing the core concentration decreased the wettability due to the presence of increased amount of surface oil present on the powder particles as stated in the previous section. Powders with higher amounts of unencapsulated oil on the surface tend to stick together and form lumps and hence reduced the wettability (Konstance *et al.*, 1995: Vega *et al.*, 2005).

A second order non-linear regression equation was fitted between dependent and independent variables using the experimental values. Following regression models were obtained to predict the bulk density of microencapsulated cumin oil powder (4.7) and naoencapsulated cumin oil powder (4.8).

$$\text{Powder wettability} = 188.24+49.01A+12.60B+38.11C+7.30AB-12.53AC +4.45BC+12.09A^2-10.73B^2+9.84C^2 \quad \text{.....(4.7)}$$

$$\text{Powder wettability} = 226.64+64.81A+19.38B+47.21C+10.92AB-3.30AC +5.78BC+21.45A^2-14.02B^2+19.36C^2 \quad \text{.....(4.8)}$$

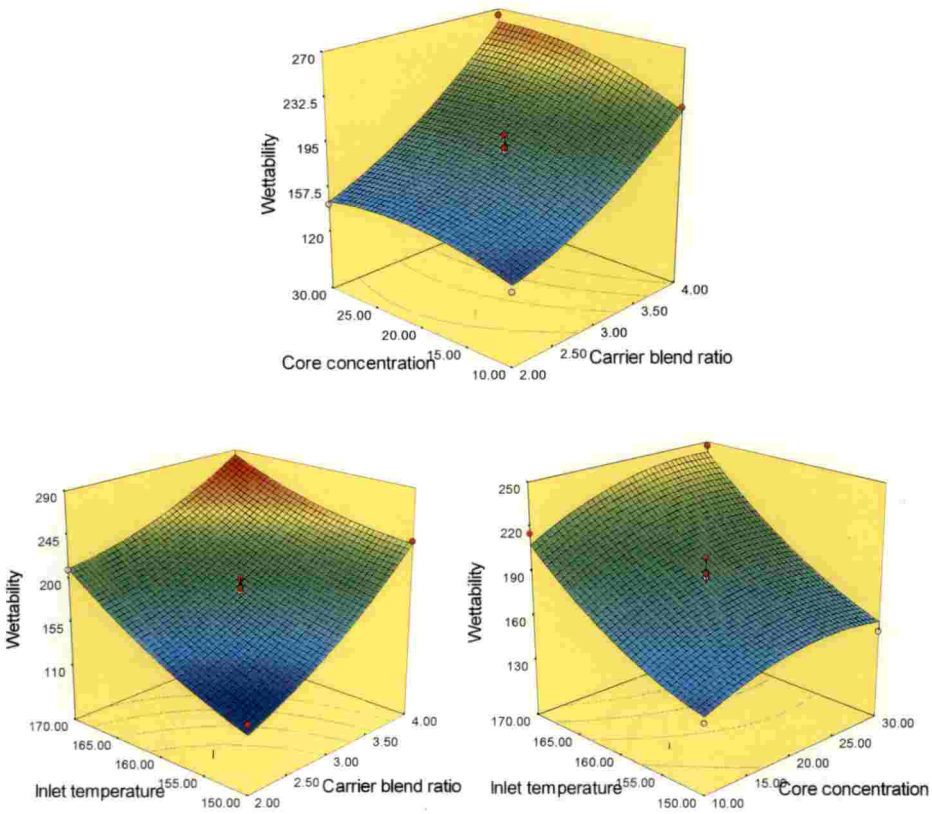


Fig. 4.7 Effect of process variables on wettability of microencapsulated cumin oil powder

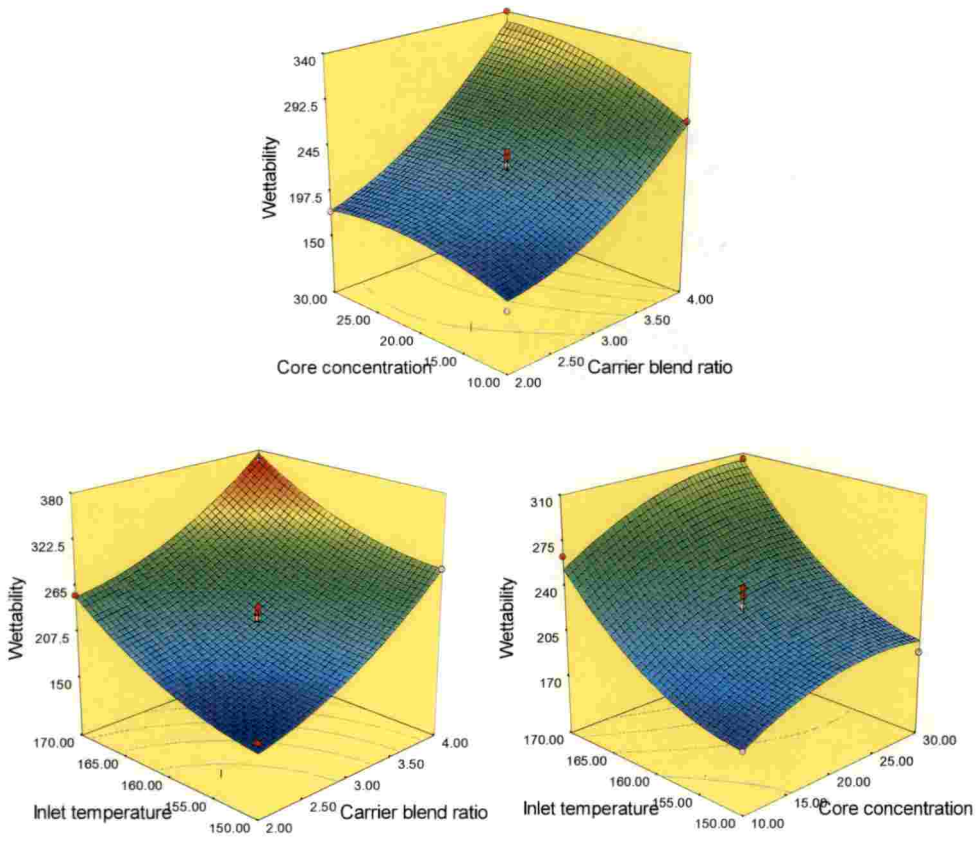


Fig. 4.8 Effect of process variables on wettability of nanoencapsulated cumin oil powder

ANOVA performed to evaluate the significance of the coefficients of the quadratic polynomial models is given in Appendix A (Table A8) and B (Table B8) for microencapsulated and nanoencapsulated cumin oil powders respectively. For the wettability of microencapsulated cumin oil powder, A, B, C, AC and A^2 were significant model terms. The p-value of the model was also less than 0.0001. From Table A8, the value of R^2 was 0.9794.

In case of wettability of nanoencapsulated cumin oil powder, A, B, C, A^2 and C^2 were significant model terms. The p-value of the model was less than 0.0001, which indicated that the model fitness was significant. From Table B8, R^2 was noted as 0.9828.

4.2.1.5 Cold Water Solubility

Solubility is considered as an important property of the powder used as ingredients for the food industry and they must exhibit good solubility. Solubility is the last particle dissolution step and is a decisive factor for the quality of these products (Jayasundera *et al.*, 2011). In the present work, all the powders were relatively soluble despite the hydrophobic nature of the core material (cumin oil).

The effect of the three independent process variables *viz.*, core concentration, carrier blend ratio and spray dryer inlet temperature on the cold water solubility of microencapsulated and nanoencapsulated cumin oil powders produced with different treatments are tabulated in Appendix A (Table A9) and B (Table B9) respectively.

The cold water solubility of microencapsulated cumin oil powder varied between 87.55 to 94.89%. In this case, the maximum value of cold water solubility was obtained for the powder produced from a gum arabic:maltodextrin ratio of 1:2, 30% core concentration and a spray dryer inlet temperature of 160°C. The minimum value was recorded for the powder produced with a gum arabic:maltodextrin ratio of 1:4, 20% core concentration and at 150°C spray dryer inlet temperature. In case of nanoencapsulation of cumin oil the values of cold water solubility varied between 91.45 and 97.77%. The maximum and minimum value was recorded for the powder produced as in case of microencapsulation.

The effect of process variables on solubility of microencapsulated and nanoencapsulated cumin oil powders are shown by plotting 3D graphs representing the response surface generated by the model (Equation. 4.9 and 4.10) and is shown in Fig.4.9 and 4.10.

The microencapsulation process facilitates the solubility of the essential oils in water by preventing phase separation (Botrel *et al.* 2012).

From the Fig.4.9 and 4.10 it is clear that the solubility is affected by the wall materials used. In the present study it was observed that the solubility decreased when the amount of maltodextrin is increased in the emulsion. This might be due to the fact that, when the gum arabic proportion in the emulsion increases, the viscosity of the emulsion increases. This will result in the formation of small sized powder particles and an increased surface area contributing to increased solubility (Prince, *et al.*, 2014).

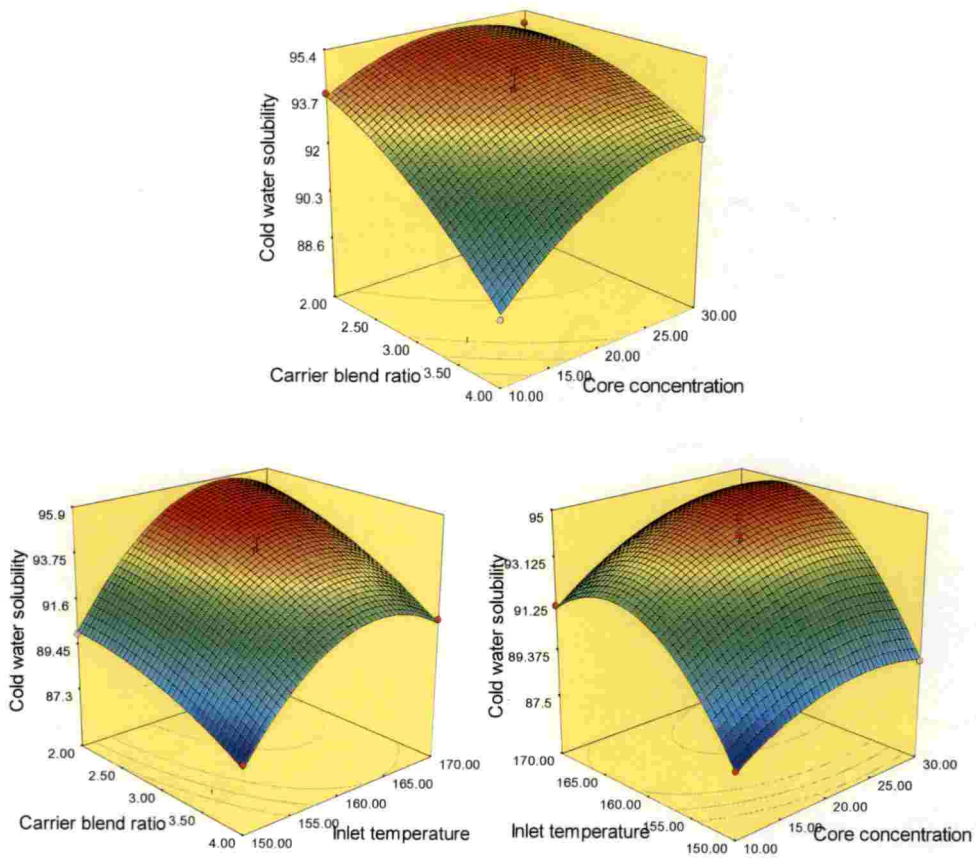


Fig. 4.9 Effect of process variables on cold water solubility of microencapsulated cumin oil powder

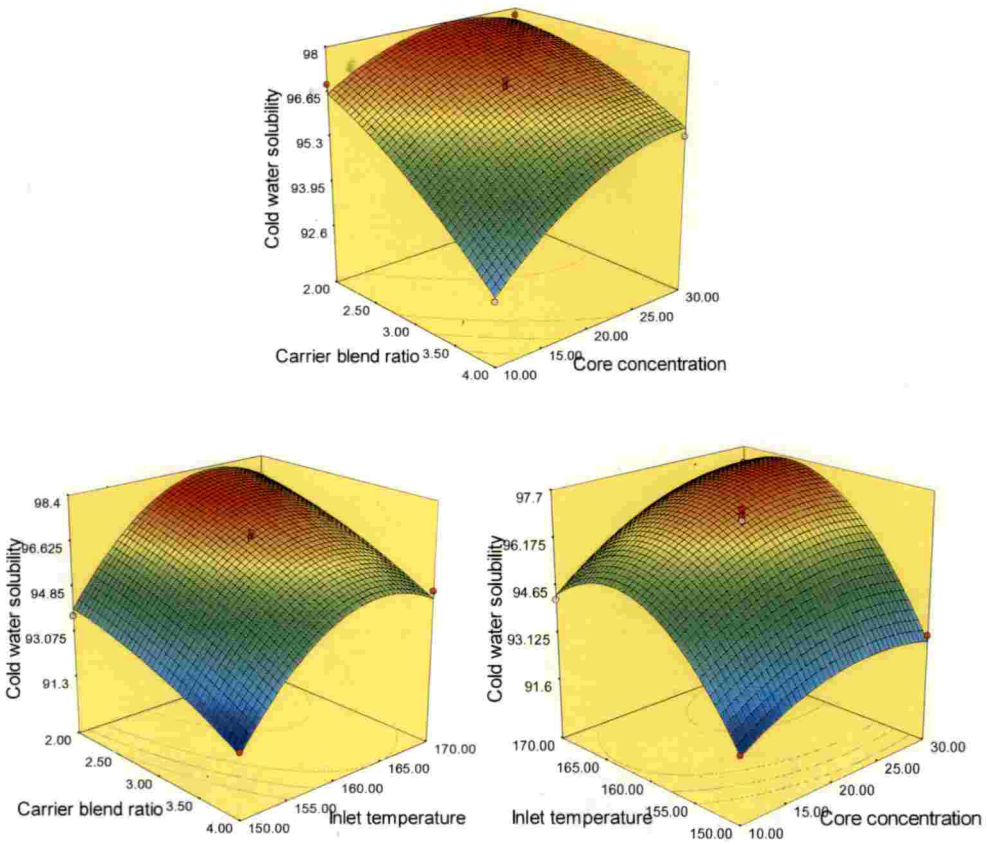


Fig. 4.10 Effect of process variables on cold water solubility of nanoencapsulated cumin oil powder

The solubility of the encapsulated powders increased with an increase in the inlet air temperature. The reason may be that when the emulsion get heated in the spray dryer, the tight organization of starch granules get disrupted, which will facilitate the migration of water into the granules and additional leaching out of soluble components (Loksuwan, 2007). In addition to this, with the increase in inlet air temperature, the amyl pectin branched chain length could have broken contributing to the increased solubility (Sariga and Prince, 2016).

The cold water solubility of both encapsulated powders were also affected by the core concentration. It was observed that when the core concentration was increased in the emulsion, there was an increase in the solubility of the powder. This may be due to that the hydrophilic wall

material available would be limited to produce a strong structural matrix resulting in thinner layers of wall material between encapsulated oil droplets (Mc Name *et al.*, 1998; Sariga and Prince, 2016).

A second order non-linear regression equation was fitted between dependent and independent variables using the experimental values. Following regression models were obtained to predict the bulk density of microencapsulated cumin oil powder (4.9) and nanoencapsulated cumin oil powder (4.10).

Cold water solubility = 94.07-1.73A+1.16B+2.14C+0.70AB-0.36AC

$$+0.37BC-0.61A^2-0.98B^2-2.57C^2 \quad \text{.....(4.9)}$$

Cold water solubility = 96.85-1.41A+1.00B)+1.74C+0.49AB-0.065AC

$$+0.38BC-0.37A^2-0.77B^2-2.05C^2 \quad \text{.....(4.10)}$$

ANOVA performed to evaluate the significance of the coefficients of the quadratic polynomial models is given in Appendix A (Table A10) and B (Table B10) for microencapsulated and nanoencapsulated cumin oil powders respectively. For the cold water solubility of microencapsulated cumin oil powder, A, B, C, AB, A^2, B^2 and C^2 were significant model terms. Among all the significant model terms A, B, C and C^2 were highly significant with a p-value less than 0.0001. The p-value of the model was less than 0.0001, which indicated that the model fitness was significant. The value of coefficient of determination (R^2) of the regression model implies that the model could account 99.22% variability in data.

In case of solubility of nanoencapsulated cumin oil powder, A, B, C, AB, BC, A^2, B^2 and C^2 were significant model terms. The p-value of the model was less than 0.0001, which indicated that the model fitness was significant. From Table B10, the value of R^2 was 0.9927 which implies that the model could account 99.27% variability in data.

4.2.1.6 Colour

Whiteness of the powder is one of the important quality parameters when it comes to food applications. Higher the values of L*, the more the white colour for the encapsulated powder. Hence in the present study, the effect of carrier blend ratio, core concentration and spray dryer inlet temperature on the Hunter 'L*' values were taken into account.

The effect of the three independent process variables on the L* value of microencapsulated and nanoencapsulated cumin oil powders produced with different treatments are tabulated in Appendix A (Table A11) and B (Table B11) respectively.

The L* value of microencapsulated cumin oil powder varied between 84.85 and 88.92. For the microencapsulated cumin oil powder, the maximum value of L* was obtained for the powder produced with gum arabic:maltodextrin ratio of 1:4, 20% core concentration and a spray dryer inlet temperature of 150°C. The minimum value was recorded for the powder produced with a gum arabic:maltodextrin ratio of 1:2, 20% core concentration and at 170°C spray dryer inlet temperature. In case of nanoencapsulated cumin oil powder, the values of L* varied between 87.69 and 91.80. The maximum and minimum values were obtained for the same treatment as in case of microencapsulation.

The effect of gum arabic:maltodextrin ratio, core concentration and spray dryer inlet temperature on L* value of microencapsulated and nanoencapsulated cumin oil powders are shown by plotting 3D graphs representing the response surface generated by the model (Equation. 4.11 and 4.12) and are shown in Fig.4.11 and 4.12.

From the figures it may be observed that, when the proportion of maltodextrin in the emulsion was increased, the L* value of both the encapsulated powders increased significantly. This may be due to higher L*

value of maltodextrin than gum arabic. A reduction in the degree of whiteness in the present study was occurred due to the increasing amount of gum arabic encapsulant in the emulsion. This may be due to the yellow-white colour of gum arabic (Pratiwi *et al.*, 2016).

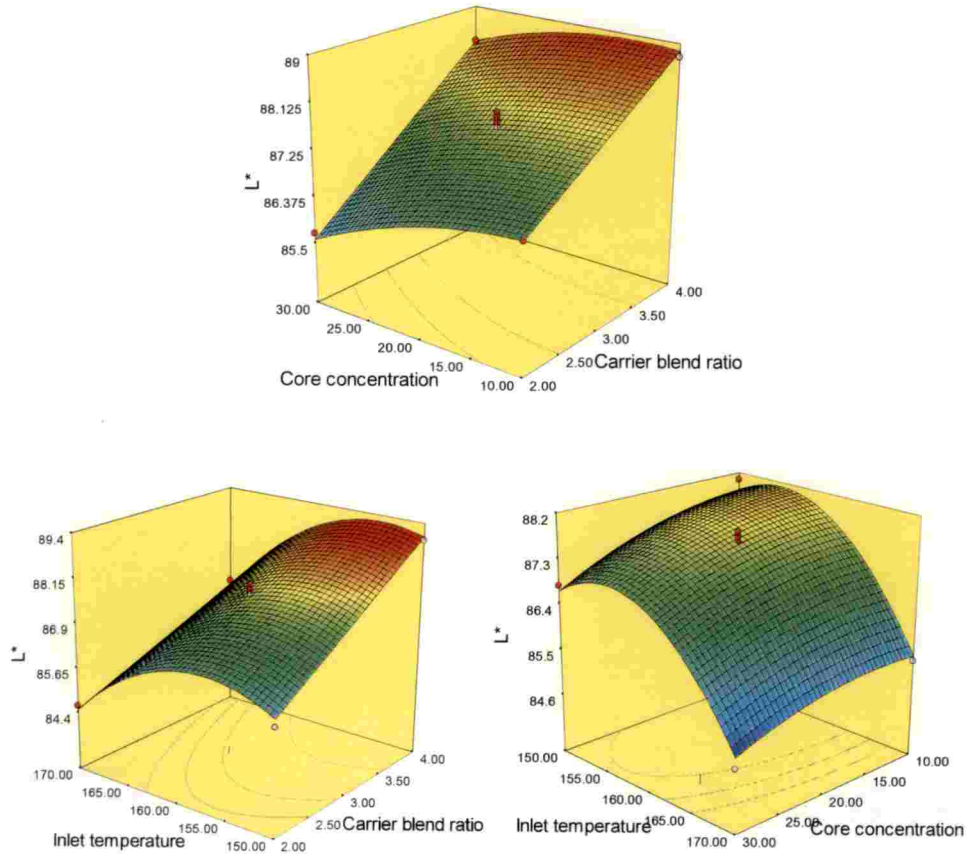


Fig. 4.11 Effect of process variables on colour of microencapsulated cumin oil powder

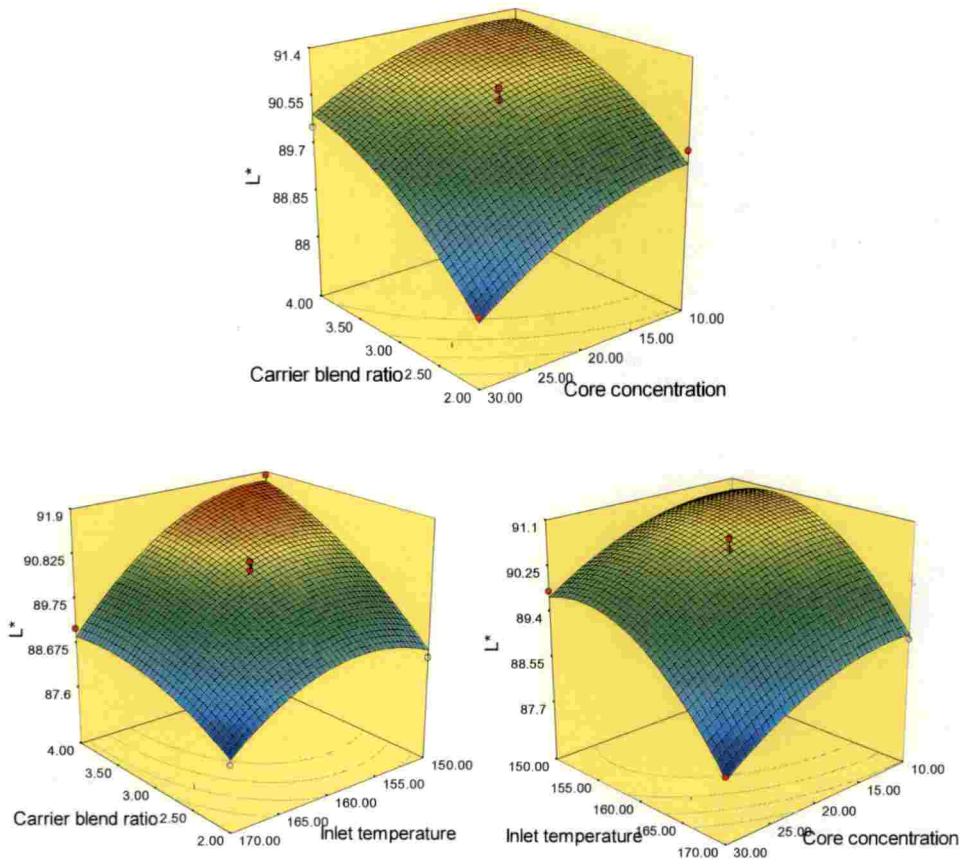


Fig. 4.12 Effect of process variables on colour of nanoencapsulated cumin oil powder

The figures indicate that the L* value of the encapsulated powder were affected by the spray dryer inlet temperature also. It was observed that the L* value of the powder were decreased when the inlet temperature was increased from 150 to 170°C. It may be due to caramelisation and Millard reactions that occurred in the presence of sugars at high temperature during drying (Lee *et al.*, 2017).

The L* value of the encapsulated powders were also affected by the core concentration. When the concentration of the oil was increased in the emulsion the L* value was found to decrease. This is due to the fact that the L* value of the cumin oil is very less.

A second order non-linear regression equation was fitted between dependent and independent variables using the experimental values. The following regression models were obtained to predict the bulk density of microencapsulated cumin oil powder (4.11) and naoencapsulated cumin oil powder (4.12).

$$L^* = 87.62+1.22A-0.43B-1.05C+0.15AB-0.31AC+0.14BC-7.500E-003A^2 -0.24B^2-1.17C^2 \quad \text{.....(4.11)}$$

$$L^* = 90.52+0.97A-0.63B-0.95C+0.14AB-0.45AC-0.043BC-0.40A^2 -0.36B^2-0.82C^2 \quad \text{.....(4.12)}$$

ANOVA performed to evaluate the significance of the coefficients of the quadratic polynomial models is given in Appendix A (Table A12) and B (Table B12) for microencapsulated and nanoencapsulated cumin oil powders respectively. For the L* value of microencapsulated cumin oil powder, A, B, C, AC and C^2 were significant model terms. The p-value of the model was less than 0.0001, which indicated that the model fitness was significant. From Table A12, it may be inferred that the value of R^2 was 0.9849 which implies that the model could account 98.49% variability in data.

In case of L* value of nanoencapsulated cumin oil powder, A, B, C, AC, A^2, B^2 and C^2 were significant model terms. The p-value of the model was less than 0.0001, which indicated that the model fitness was significant. From Table B12, the R^2 value was found to be 0.9768.

4.2.1.7 Encapsulation Efficiency

A successful encapsulation process for flavours and oils is the one which results in an encapsulated powder having minimum of surface oil content on the powder particles and maximum retention of the core material, particularly volatiles, inside the encapsulated powder particles (Jafari *et al.* 2008).

The values of encapsulation efficiency obtained from different combinations core concentration, carrier blend ratio and spray dryer inlet temperature are organised in Appendix A (Table A13) and B (Table B13) respectively.

The encapsulation efficiency of microencapsulated cumin oil powder varied between 68.56 to 93.88%. The maximum encapsulation efficiency value was obtained for the powder produced with a gum arabic:maltodextrin ratio of 1:2, 20% core concentration and a spray dryer inlet temperature of 170°C. The minimum value was recorded for the powder produced with a gum arabic:maltodextrin ratio of 1:4, 20% core concentration and at 150°C spray dryer inlet temperature. In case of nanoencapsulation of cumin oil the values of encapsulation efficiency varied between 65.35 and 89.62%. The maximum and minimum value was obtained for the same treatment as in case of microencapsulated cumin oil powder.

The effects of process variables on encapsulation efficiency are illustrated by plotting 3D graphs representing the response surface generated by the model (Equation. 4.13 and 4.14) and are shown in Fig.4.13 and 4.14.

From the results obtained in the present study, it was observed that the spray dryer inlet temperature was the most significant independent parameter that greatly affected the total oil retention. When the spray dryer inlet temperature was increased, it improved the retention of cumin oil and a decrease in oil content on the particle surface. Increasing the inlet air

temperature (within limits) will result in a rapid formation of a semipermeable membrane around the emulsion droplet, which offers better retention of volatile oil (Bringas-Lantigua *et al.*, 2011). The rapid drying rate would also probably make the crust layer more firm and no further leaching of the volatile would occur towards the surface (Bhandari, *et al.* 1992). The same results were also reported by Bringas-Lantigua *et al.* (2011) in microencapsulation of mandarin oil by spray drying, Bringas-Lantigua *et al.* (2012) in microencapsulation of lime essential oil and Tonon *et al.* (2011) in microencapsulation of flaxseed oil.

From Fig. 4.13 and 4.14, it is clear that there is a decrease in volatile retention when the ratio of gum arabic:maltodextrin in the emulsion was increased from 1:2 to 1:4 and this is justified by gum arabic's better film forming and improved emulsifying/stabilising properties (Madene *et al.*, 2006). The results are in consistent with those of Fernandes *et al.* (2008a) in microencapsulation of *Lippia sidoides* essential oil. In this study maltodextrin offered the lowest volatile oil retention. The reason for this may be explained by their poor film – forming ability. Maltodextrin have low emulsification properties and hence they will produce coarse emulsions and poor flavour retention during drying. Badee *et al.* (2012) and Shaikh *et al.* (2006) also reported that gum arabic provides increased volatile oil retention.

Results of the present research work indicated that when the core concentration was increased or there was higher oil content in the emulsions, the amount of surface oil or non encapsulated oil was increased with less oil being encapsulated. The same results have also been obtained by Dian *et al.* (1996) in microencapsulation of palm-based oil and Badee *et al.* (2012) in microencapsulation of peppermint oil. The reason for this might the instability of the oil droplets in the emulsion before the spray drying process. This instability results in the leakage of the oil onto the surface of the microcapsules during the drying process (Faldt and

Bergenstahl, 1995). It may also be due to the presence of insufficient wall material for encapsulating the oil, as a result of increased oil content in the emulsion. Carmona *et al.* (2013) also reported the same result that oil concentration showed a positive effect on the surface oil, i.e., higher oil concentrations led to higher surface oil in the microencapsulation of orange essential oil. This can also be due to greater amounts of cumin oil close to the drying surface, which will make the diffusion path length short to the air/particle interface, thereby increasing the amount of surface oil (Badee *et al.*, 2012).

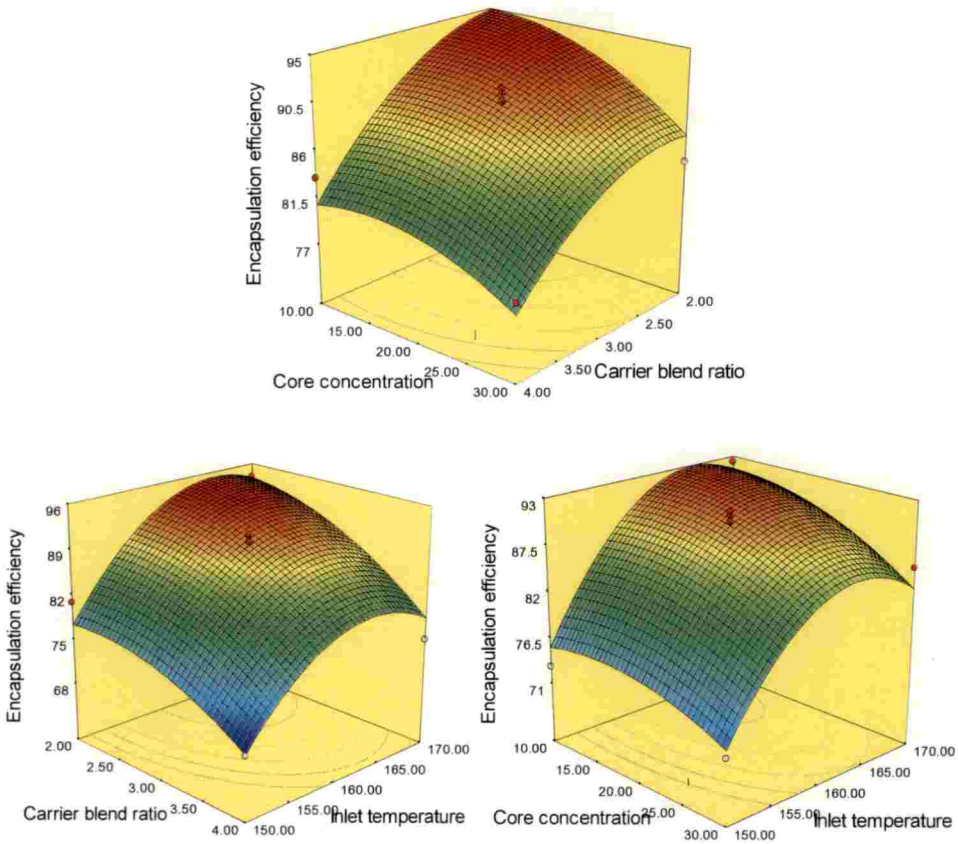


Fig. 4.13 Effect of process variables on encapsulation efficiency of microencapsulated cumin oil powder

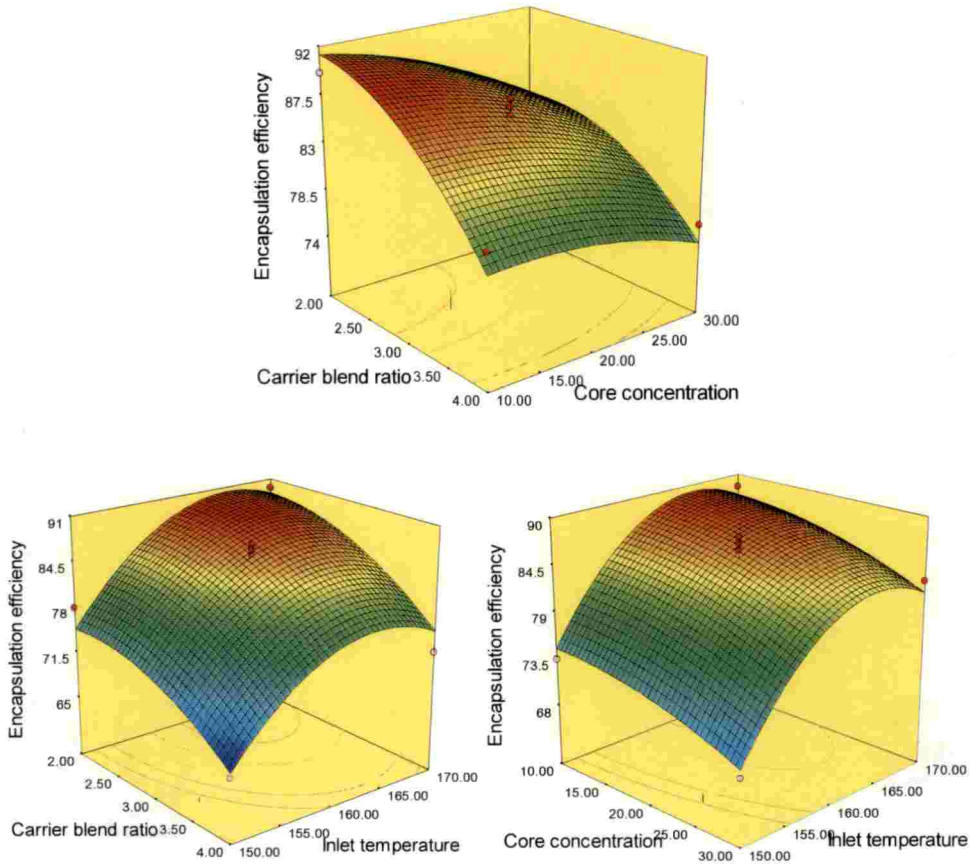


Fig. 4.14 Effect of process variables on encapsulation efficiency of nanoencapsulated cumin oil powder

A second order non-linear regression equation was utilised to relate between dependent and independent variables using the experimental values. Following regression models were obtained to predict the encapsulation efficiency of microencapsulated cumin oil powder (4.13) and nanoencapsulated cumin oil powder (4.14).

$$\text{Encapsulation efficiency} = 89.96 - 5.87A - 2.92B + 6.62C + 1.15AB - 1.47AC - 1.35BC - 2.89A^2 - 2.11B^2 - 7.33C^2 \quad \text{.....(4.13)}$$

$$\text{Encapsulation efficiency} = 86.08 - 5.33A - 2.89B + 6.10C + 1.17AB - 0.87AC - 0.22BC - 2.92A^2 - 1.27B^2 - 6.55C^2 \quad \text{.....(4.14)}$$

ANOVA performed to evaluate the significance of the coefficients of the quadratic polynomial models is given in Appendix A (Table A14) and B (Table B14) for microencapsulated and nanoencapsulated cumin oil powders. In case of encapsulation efficiency of microencapsulated cumin oil powder, A, B, C and C^2 were significant model terms. The p-value of the model was 0.0016, which indicated that the model fitness was significant. From Table A14, the value of coefficient of determination R^2 was 0.9410 and it implies that the model could account 94.10% variability in data.

For the encapsulation efficiency of nanoencapsulated cumin oil powder, A, B, C and C^2 are significant model terms. The p-value of the model was 0.0011, which indicated that the model fitness was significant. The value of coefficient of determination (R^2) of the regression model implies that the model could account 94.74% variability in data (Table B14).

4.3 PROCESS OPTIMISATION FOR MICROENCAPSULATED AND NANOENCAPSULATED CUMIN OIL POWDERS

The microencapsulation and nanoencapsulation of cumin oil was carried out using the spray drying method and the various process variables which will affect the quality characteristics of the encapsulated powders were studied. Optimisation of the three independent variables such as carrier blend ratio (gum arabic:maltodetrin in 1:2, 1:3 and 1:4), core concentration (10, 20 and 30%) and spray dryer inlet temperature (150, 160 and 140°C) were done by Box- Behnken design of response surface methodology in Design Expert Software 7.7.0. and the desirability analysis was performed using this software. Desirability ranges from zero to one for any given response. A zero indicates that one or more responses fall outside desirable limits and a value of one represents the ideal case (Maran *et al.*, 2013). In this study, the independent variables were kept within the range and the dependent variables were selected as minimum or maximum depending

upon the requirements. From the desirability analysis, the optimal level of various parameters were found and listed in Table 4.4 and 4.5. The optimum operating conditions for microencapsulation of cumin oil was found as a carrier blend ratio (gum arabic:maltodextrin) of 1: 2.77, core concentarion of 10% and a spray dryer inlet temperature of 162.50°C whereas for nanoencapsulation it was found to be a carrier blend ratio of 1:2.92, core concentration of 10% and a spray dryer inlet temperature of 163.38°C. The desirability of the optimisation of microencapsulation of cumin oil was found to be 0.704 and that of nanoencapsulation was 0.717. Since these desirability values are closer to one, the optimised values could be considered ideal.

Table 4.3 Optimal level obtained for microencapsulated cumin oil powder from the desirability analysis

Sl. No.	Response	Desirability	Optimal level	Lower limit	Upper limit
1	Carrier blend ratio	Is in range	2.77	2	4
2	Core concentration	Is in range	10.00	10	30
3	Inlet temperature	Is in range	162.50	150	170
4	Moisture content	Minimise	4.33	3.68	5.55
5	Bulk density	Maximise	0.425	0.317	0.47
6	Wettability	Minimise	165.53	120.3	275
7	Colour	Maximise	87.21	84.59	88.92
8	Water activity	Minimise	0.31	0.261	0.385
9	Encapsulation efficiency	Maximise	93.88	68.56	93.88
10	Cold water solubility	Maximise	92.77	87.55	94.95

Table 4.4 Optimal level obtained for nanoencapsulated cumin oil powder from the desirability analysis

Sl. No.	Response	Desirability	Optimal level	Lower limit	Upper limit
1	Carrier blend ratio	Is in range	2.92	2	4
2	Core concentration	Is in range	10.00	10	30
3	Inlet temperature	Is in range	163.37	150	170
4	Moisture content	Minimise	3.24763	2.7	4.23
5	Bulk density	Maximise	0.454858	0.37	0.495
6	Wettability	Minimise	205.196	150.2	364.8
7	Colour	Maximise	90.3258	87.69	91.8
8	Water activity	Minimise	0.239672	0.205	0.338
9	Encapsulation efficiency	Maximise	89.6239	65.35	89.62
10	Cold water solubility	Maximise	95.4692	91.45	97.77

4.4 PARTICLE SIZE ANALYSIS OF MICROENCAPSULATED AND NANOENCAPSULATED CUMIN OIL POWDERS

The particle size of the optimised microencapsulated and nanoencapsulated cumin oil powders were determined using Zetasizer nano range. The particle size of the microencapsulated cumin oil powder was found to be 3.033μm and that of nanoencapsulated cumin oil powder was 63.47 nm. The results obtained for microencapsulated and nanoencapsulated powders were in their respective ranges.

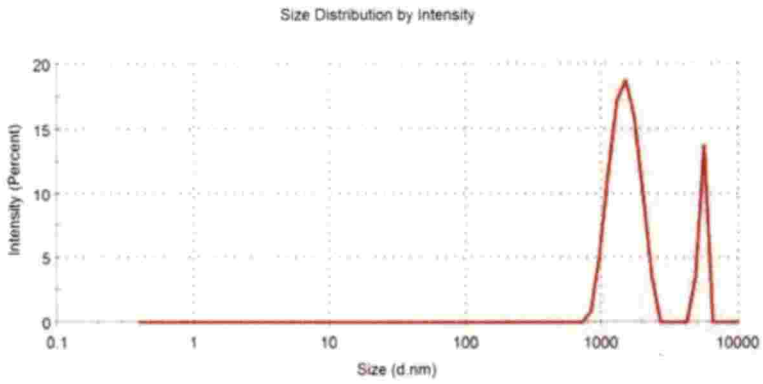


Fig. 4.15 Particle size of microencapsulated cumin oil powder

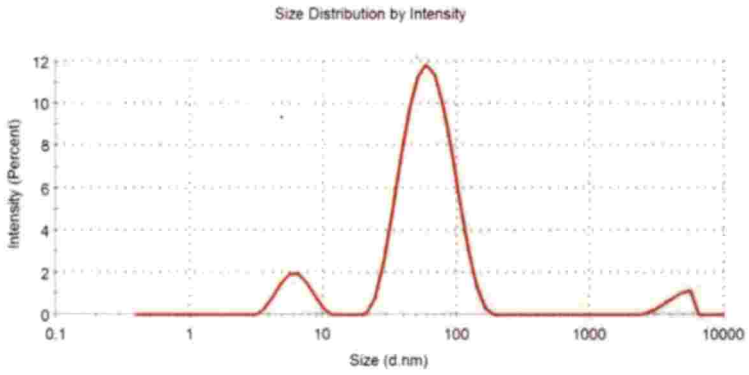


Fig.4.16 Particle size of nanoencapsulated cumin oil powder

120

4.5 MORPHOLOGICAL CHARACTERISTICS OF THE MICROENCAPSULATED AND NANOENCAPSULATED CUMIN OIL POWDERS

The morphological studies of optimally produced microencapsulated and nanoencapsulated cumin oil powders were carried out using Scanning Electron Microscopy (SEM). The SEM micrographs of morphology of microencapsulated and nanoencapsulated cumin oil powders are shown in Fig. 4.17 and 4.18. From the SEM micrographs it was observed that in general both the microencapsulated and nanoencapsulated capsules obtained were smooth but not completely spherical with the presence of surface dents which may probably formed by shrinkage of the droplets during the early stages of the drying process (Re, 1998). The outer surface of both the powders was free of cracks. The presence of surfaces without apparent breakage or fissures is important because any damage in particle surface could reduce the protection and retention of the core compound (Trindade and Grosso, 2000; Porras-Saavedra *et al.*, 2015) and the results indicated that the powder had low permeability to gases and better protection and retention of the cumin oil.

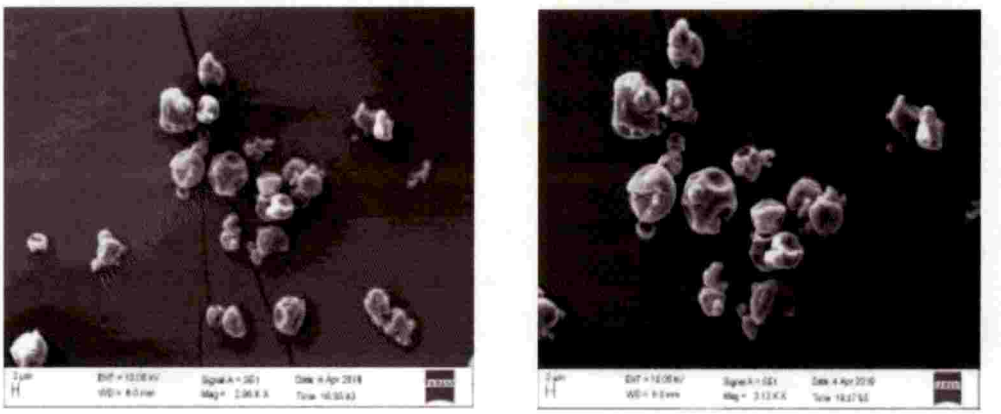


Fig.4.17 SEM micrographs of optimally produced microencapsulated cumin oil powder

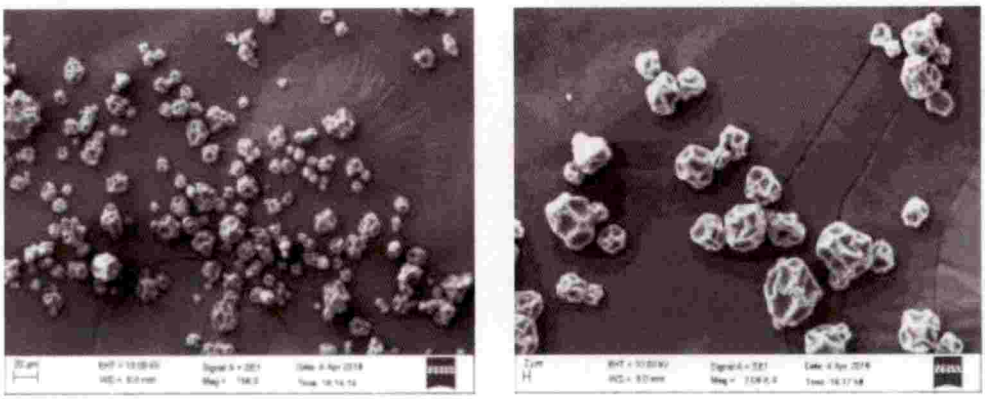


Fig.4.18 SEM micrographs of optimally produced nanoencapsulated cumin oil powder

4.6 RETENTION OF ACTIVE COMPONENTS IN MICROENCAPSULATED AND NANOENCAPSULATED CUMIN OIL POWDERS

The main active components of cumin oil are cuminaldehyde and γ-terpinene. The presence these two components in the optimised microencapsulated and nanoencapsulated cumin oil powders were determined using Gas Chromatography (GC) method. The presence of these two components were analysed by comparing with the chromatograph of cuminaldehyde and γ- terpinene standards. The gas chromatographs of cuminaldehyde and γ- terpinene standards are given in Fig.4.19 and 4.20 respectively. The obtained gas chromatographs of the optimally produced microencapsulated and nanoencapsulated cumin oil powders are shown in Fig. 4.21 and 4.22 respectively.

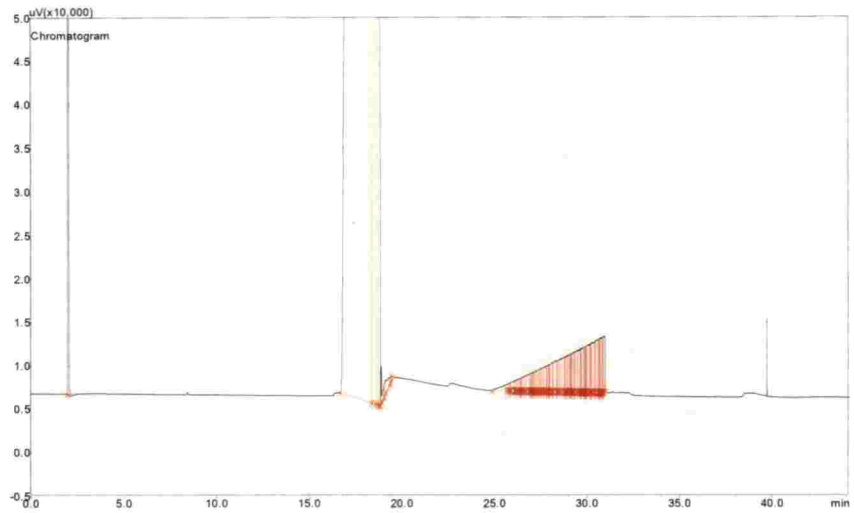


Fig.4.19 Gas chromatograph of cuminaldehyde standard

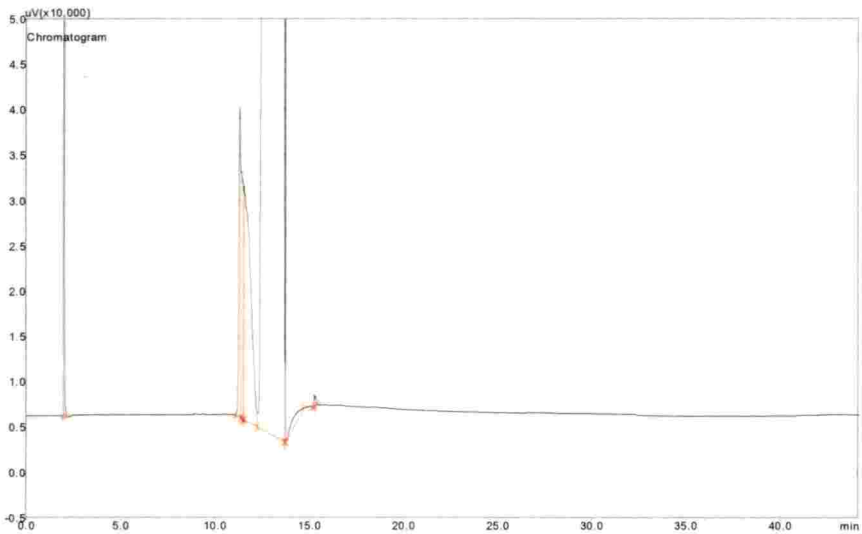


Fig.4.20 Gas chromatograph of γ- terpinene standard

123

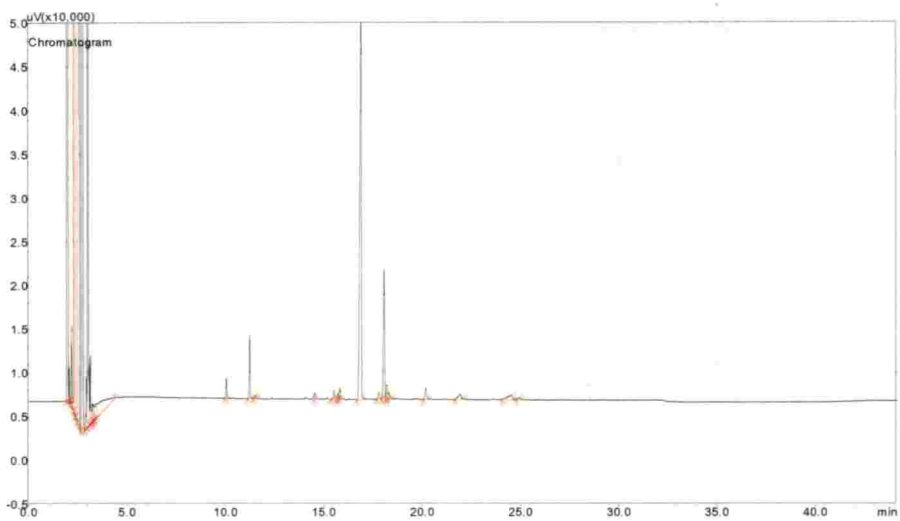


Fig.4.21 Gas chromatograph of microencapsulated cumin oil

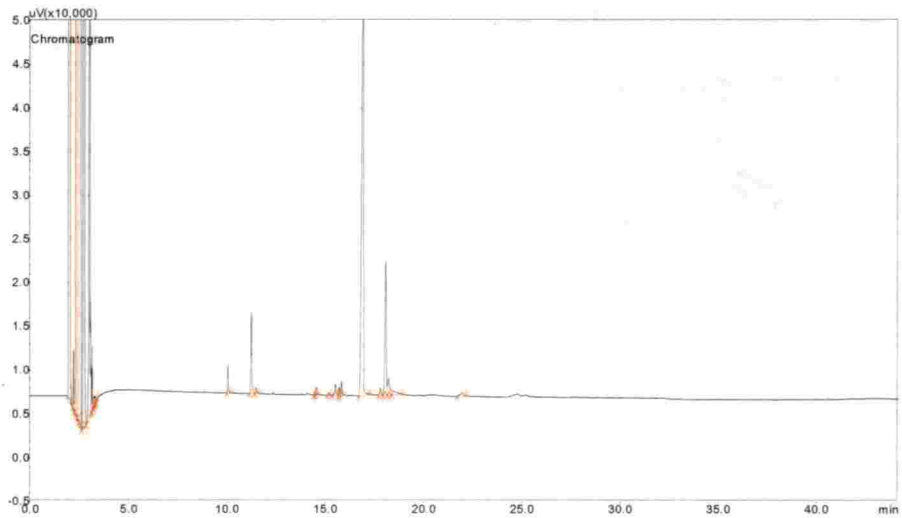


Fig.4.22 Gas chromatograph of nanoencapsulated cumin oil

It is clear from the figures that the peaks of cuminaldehyde and γ-terpinene in the chromatographs of the optimised samples were similar to those obtained for their respective standards and which illustrated the presence of both the components in the microencapsulated and nanoencapsulated cumin oil powders.

4.7 COMPARISON OF OPTIMALLY PRODUCED MICROENCAPSULATED AND NANOENCAPSULATED CUMIN OIL POWDERS

The results obtained in the present work showed that moisture content and water activity of microencapsulated cumin oil powder were higher than that of nanoencapsulated powder. The reason for this may be due to the reduction in the size of atomised emulsion droplets in nanoencapsulation which would greatly increase the contact surface for the heat and mass transfer. As a result of this severe heat and mass transfer, the water vapourisation from droplet/particle is accelerated and led to particles with lower moisture content. These findings are similar to those obtained by Aghbashlo *et al.* (2013) in fish oil microencapsulation.

In principle, density increases as volume decreases at a given constant mass. From a theoretical point of view, particles having smaller diameter contribute to a tighter packing and thereby results in an increased bulk density. In the present study, the bulk density of nanoencapsulated cumin oil powder was found to be higher than that of the microencapsulated cumin oil powder.

The wettability decreased with the decrease in particle size. This may be due to the lower moisture content of the powder obtained in nanoencapsulated powder particles (Fernandes *et al.*, 2013). Caking, usually occurs in powder particles containing higher amount of moisture and these moisture can contribute to the wettability since the liquid penetrates into the pores more easily (Buffo *et al.*, 2002; Fernandes *et al.*, 2013). A caked

product allows the larger particles to settle to the bottom and disperse more easily in water.

A decrease in particle size contributes to increased surface area contributing to increased solubility (Prince *et al.*, 2014) and hence nanoencapsulated powder had higher solubility than microencapsulated powder.

The small particles normally produced by spray drying are largely invisible as individual particles and have a light colour. Agglomerated or larger powder particles appear as discernable particles and are darker and richer in colour. The L* values were higher for the nanoencapsulated powder than the microencapsulated powder indicating they are whiter. Thus, any factors that have been mentioned as influencing particle size, including agglomeration, also influence appearance of the powder (Reineccius 2004).

In case of encapsulation efficiency, the microencapsulated powder showed higher values than the nanoencapsulated cumin oil powder. This may due to the reason that the smaller particles possess increased surface area to volume ratio. Hence they contain more surface oil than the microencapsulated powder, which would have resulted in reduced flavour retention. The most important process parameter in determining the volatiles retention and encapsulation efficiency of the oils during spray drying is the dissolved solids content in the feed. When high solid contents are used in the emulsion, the volatile retention is increased principally by reducing the required time for the formation of a semi-permeable membrane at the surface of the drying droplets. Also higher total solids increase the emulsion viscosity thereby preventing the circulation movement inside the droplets and results in a rapid skin formation (Fernandes *et al.*, 2008b; Jafari *et al.*, 2008).

The particle size obtained for both microencapsulated and nanoencapsulated powders were in their respective ranges. The SEM analysis showed that the surface dents were more in nanoencapsulated

powder than the microencapsulated powder. This can be linked to the fact that the nanoparticles undergo very fast drying (and hence early solidification) as a result of high surface area to volume ratio. In such situations, dents could not be "erased" by the thermal expansion of either the air or water vapours inside the drying particle.

4.8 COST ANALYSIS

The computation of cost of production of one kilogram of microencapsulated and nanoencapsulated cumin oil powders are shown in Appendix C. The computed cost of one kilogram of microencapsulated cumin oil powder using twin fluid nozzle was found to be Rs. 1577/kg and that for nanoencapsulated cumin oil powder using ultrasonic atomiser was found to be Rs. 13510.7/kg. The cost of the production could be further scaled down once the production is taken up on a commercial scale.

Summary and Conclusion

CHAPTER V

SUMMARY AND CONCLUSION

Spices are the main flavouring agents in food and were in use since the birth of civilisation. It is mainly used for providing aroma, taste, flavour, texture and colour to food, and also as preservative and can provide nutritional, and health benefits. Cumin (*Cuminum cyminum Linn.*) is one of the important commercial seed spices and is valued for its aroma, medicinal and therapeutic properties. In India, the area under cumin cultivation in the year 2015-2016 was 808230 ha and the production was 503260 tonnes. The main cumin cultivating states are Rajasthan, Gujarat and Madhya Pradesh.

Essential oil content is the most important chemical constituent of cumin seeds which ranges from 2.5 to 4.5% and is pale to colourless depending on age and regional variations. This oil obtained by steam distillation is used to flavour alcoholic beverages, desserts and condiments. In addition, it can be used as a fragrant component of creams, lotions and perfumes.

The biological activity of the oil is lost due to the volatilisation or degradation of active compounds when they are exposed to heat, humidity, light, or oxygen. Hence these oils have to be protected from all these factors. Encapsulation is the most suitable method which can be used to protect the cumin oil from the liquid form to solid form by coating agents. Microencapsulation and nanoencapsulation are the two encapsulating techniques commonly used. If the encapsulated product have sizes in micrometre or larger, then it is called microencapsulation whereas nanoencapsulation is the application of encapsulation in the nanometre scale. Spray drying is the most extensively used encapsulation technique in the food industry. It is a cost-effective, rapid, continuous, scalable and reproducible process used for producing dry powders from fluid materials.

The encapsulation of cumin oil could be carried out with the selection of suitable wall materials. The intention of the present study was to optimise the process parameters for the microencapsulation and nanoencapsulation of cumin oil by spray drying technique.

The microencapsulation and nanoencapsulation of cumin oil was carried out using spray dryer with twin fluid atomiser and ultrasonic atomiser respectively. Suitable wall materials were selected by conducting preliminary studies with gum arabic, maltodextrin and tapioca starch. The results of preliminary studies showed that tapioca starch could not significantly encapsulate cumin oil and hence it was eliminated. Combinations of gum arabic and maltodextrin were selected as the suitable wall materials for the encapsulation of the cumin oil as they could significantly encapsulate cumin oil. The physico-chemical properties of cumin oil and wall materials were found out. Encapsulation was carried out with three different carrier blend ratio (1:2, 1:3 and 1:4), core concentration (10, 20 and 30%) and spray dryer inlet temperature (150, 160 and 170°C).

Initially the emulsion was prepared by mixing the wall materials and the cumin oil with a high speed mixture and the emulsion was fed to the spray dryer. After spray drying, the encapsulated cumin oil powder were collected and packed in aluminum foil pouches. The quality characteristics of the encapsulated powders such as moisture content, water activity, bulk density, wettability, colour, cold water solubility and encapsulation efficiency were determined by standard methods and the optimisation was done with Box-Behnken design in RSM (Response Surface Methodology).

The moisture content of microencapsulated cumin oil powder varied from 3.68 to 5.55% (w.b.) whereas for nanoencapsulation it was from 2.7 to 4.23 %. The moisture content showed a decreasing tendency when the spray dryer inlet temperature increased from 150 to 170°C and proportion of maltodextrin was increased from 1:2 to 1:4. The core concentration did not

show any effect on the moisture content. The minimum moisture content was observed for the powder produced from a gum arabic:maltodextrin ratio of 1:4, 20% core concentration and at 170°C spray dryer inlet temperature.

The water activity of microencapsulated cumin oil varied between 0.261 and 0.385 whereas in case of nanoencapsulated cumin oil powder it varied between 0.205 and 0.338. The effect of process variables on water activity was same as that of the moisture content. Water activity decreased with increase in moisture content and the spray dryer inlet temperature. The core concentration did not significantly affect the water activity. In both cases the minimum value of water activity was recorded for the samples produced from 1:4 ratio of gum arabic:maltodextrin, 20% core concentration and at 170°C.

The bulk density of microencapsulated and nanoencapsulated powder varied from 0.317 to 0.470 g/ cm^3 and 0.370 to 0.495 g/cm^3 respectively. As the inlet temperature, proportion of maltodextrin and core concentration was increased, the bulk density decreased. In case of microencapsulated cumin oil, the highest bulk density value was obtained for the powder produced with gum arabic:maltodextrin ratio of 1:2, 20% core concentration and a spray dryer inlet temperature of 150°C whereas in nanoencapsulated powder, the highest bulk density was recorded for those produced from a gum arabic:maltodextrin ratio of 1:2, 10% core concentration and a spray dryer inlet temperature of 160°C.

The wettability of microencapsulated powder varied from 120.3 s to 275 s and that for nanoencapsulated powder varied from 150.2 and 364.8 s. An increase in the inlet air temperature and decrease in gum arabic proportion resulted in an increase in average time to wet while increasing the core concentration decreased the wettability. In case of microencapsulated powder, the highest wettability was obtained for the powder produced with gum arabic:maltodextrin ratio of 1:2, 20% core

concentration and a spray dryer inlet temperature of 150°C. For the nanoencapsulated powder, the highest wettability was obtained for those produced from 1:2 ratios of carrier blends, 10% core concentration and at 160°C.

The cold water solubility of microencapsulated cumin oil powder varied between 87.55 to 94.89% and that for nanoencapsulated powder varied from 91.45 and 97.77%. An increase in the inlet temperature and core concentration increased the cold water solubility of the powder while increased amount of maltodextrin reduced the cold water solubility. For both the encapsulated powder, the maximum cold water solubility was obtained for the powder produced from a gum arabic:maltodextrin ratio of 1:2, 30% core concentration and a spray dryer inlet temperature of 160°C.

The L* value of microencapsulated cumin oil powder varied between 84.85 and 88.92 and for nanoencapsulated powder it varied between 87.69 and 91.80. When the spray dryer inlet temperature and core concentration were increased, there was a reduction in the L* value of the powder while the whiteness increased as the amount of maltodextrin in the emulsion increased. The maximum value of L* was obtained for the powder produced with a gum arabic:maltodextrin ratio of 1:4, 20% core concentration and a spray dryer inlet temperature of 150°C.

The encapsulation efficiency of microencapsulated cumin oil powder varied between 68.56 to 93.88% and that for nanoencapsulated powder varied between 65.35 and 89.62%. When the spray dryer inlet temperature was increased, the encapsulation efficiency decreased whereas an increase in core concentration and maltodextrin amount in the emulsion decreased the encapsulation efficiency. The highest encapsulation efficiency was recorded for the powder produced with a gum arabic:maltodextrin ratio of 1:2, 20% core concentration and a spray dryer inlet temperature of 170°C in both the encapsulation processes.

The optimum operating conditions for microencapsulation of cumin oil was obtained as a carrier blend ratio (gum arabic:maltodextrin) 1: 2.77, core concentration of 10% and a spray dryer inlet temperature of 162.50°C. The optimum condition for nanoencapsulation of cumin oil was found to be a carrier blend ratio of 1:2.92, core concentration of 10% and a spray dryer inlet temperature of 163.38°C.

The particle size of the optimised microencapsulated and nanoencapsulated cumin oil powders were determined using Zetasizer nano range. The particle size of the microencapsulated cumin oil powder was found to be 3.033µm and that of nanoencapsulated cumin oil powder were 63.47 nm. The morphological studies of optimally produced microencapsulated and nanoencapsulated cumin oil were carried out using Scanning Electron Microscopy (SEM). It was observed that, in general both the microencapsulated and nanoencapsulated capsules obtained were smooth but not completely spherical with the presence of surface dents. The presence the main active components *viz*., cuminaldehyde and γ- terpinene in the optimised encapsulated cumin oil powder was determined using gas chromatography method and it indicated the presence of both components in them.

The cost of production of one kilogram of microencapsulated cumin oil powder using the pilot model spray drier was estimated to be Rs. 1577/kg and that for nanoencapsulated powder it is Rs. 13510.7/kg.

The following are the suggestions for future research work on the encapsulation of cumin oil:

1. Encapsulation with different wall materials for increased encapsulation efficiency
2. Encapsulation with different methods

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CHAPTER VI

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Appendices

APPENDIX A

Table A1 Effect of process parameters on moisture content of microencapsulated cumin oil powder

Run	Carrier blend ratio	Core concentration (%)	Inlet temperature (°C)	Moisture content (w.b.) (%)
1	3	20	160	4.33
2	4	30	160	4.10
3	3	30	150	5.21
4	3	20	160	4.19
5	**4**	**20**	**170**	**3.68**
6	3	20	160	4.25
7	2	10	160	4.99
8	2	30	160	5.08
9	**2**	**20**	**150**	**5.55**
10	2	20	170	4.81
11	3	20	160	4.50
12	3	10	170	3.89
13	3	20	160	4.30
14	3	10	150	5.32
15	3	30	170	3.80
16	4	10	160	4.01
17	4	20	150	4.91

Table A2 Analysis of variance (ANOVA) for moisture content of microencapsulated cumin oil powder

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F	
Model	5.10	9	0.57	23.10	0.0002	Significant
A-Carrier blend ratio	1.74	1	1.74	70.90	< 0.0001	
B- Core concentration	5.000E-005	1	5.000E-005	2.038E-003	0.9652	
C- Inlet temperature	2.89	1	2.89	117.91	< 0.0001	
AB	0.000	1	0.000	0.000	1.0000	
AC	0.060	1	0.060	2.45	0.1617	
BC	1.000E-004	1	1.000E-004	4.077E-003	0.9509	
A^2	0.18	1	0.18	7.34	0.0302	
B^2	2.476E-003	1	2.476E-003	0.10	0.7600	
C^2	0.20	1	0.20	8.06	0.0250	
Residual	0.17	7	0.025			
Lack of Fit	0.12	3	0.039	2.87	0.1677	Not significant
Pure Error	0.055	4	0.014			
Cor Total	5.27	16				
Std. Dev.	0.16		R-Squared		0.9674	
Mean	4.52		Adj R-Squared		0.9256	
C.V.%	3.46		Pred R-Squared		0.6282	
PRESS	1.96		Adeq Precision		17.774	

Table A3 Effect of process parameters on water activity of microencapsulated cumin oil powder

Run	Carrier blend ratio	Core concentration (%)	Inlet temperature (°C)	Water activity
1	3	20	160	0.309
2	4	30	160	0.269
3	3	30	150	0.371
4	3	20	160	0.305
5	**4**	**20**	**170**	**0.261**
6	3	20	160	0.311
7	2	10	160	0.351
8	2	30	160	0.36
9	**2**	**20**	**150**	**0.385**
10	2	20	170	0.339
11	3	20	160	0.269
12	3	10	170	0.28
13	3	20	160	0.325
14	3	10	150	0.375
15	3	30	170	0.308
16	4	10	160	0.301
17	4	20	150	0.347

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Table A4 Analysis of variance (ANOVA) for water activity of microencapsulated cumin oil powder

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F	Sum of Squares
Model	0.023	9	2.503E-003	8.04	0.0059	Significant
A-Carrier blend ratio	8.256E-003	1	8.256E-003	26.52	0.0013	
B- Core concentration	1.250E-007	1	1.250E-007	4.015E-004	0.9846	
C- Inlet temperature	0.011	1	0.011	33.76	0.0007	
AB	4.203E-004	1	4.203E-004	1.35	0.2834	
AC	4.000E-004	1	4.000E-004	1.28	0.2944	
BC	2.560E-004	1	2.560E-004	0.82	0.3947	
A2	2.678E-004	1	2.678E-004	0.86	0.3846	
B2	3.024E-004	1	3.024E-004	0.97	0.3572	
C2	1.897E-003	1	1.897E-003	6.09	0.0429	
Residual	2.180E-003	7	3.114E-004			
Lack of Fit	4.387E-004	3	1.462E-004	0.34	0.8015	Not significant
Pure Error	1.741E-003	4	4.352E-004			
Cor Total	0.025	16				
Std. Dev.	0.018		R-Squared		0.9118	
Mean	0.32		Adj R-Squared		0.7983	
C.V.%	5.49		Pred R-Squared		0.6057	
PRESS	9.740E-003		Adeq Precision		10.105	

Table A5 Effect of process parameters on bulk density of microencapsulated cumin oil powder

Run	Carrier blend ratio	Core concentration (%)	Inlet temperature (°C)	Bulk density (g/cm^3)
1	3	20	160	0.35
2	4	30	160	0.368
3	3	30	150	0.467
4	3	20	160	0.35
5	**4**	**20**	**170**	**0.317**
6	3	20	160	0.338
7	2	10	160	0.469
8	2	30	160	0.431
9	**2**	**20**	**150**	**0.47**
10	2	20	170	0.419
11	3	20	160	0.352
12	3	10	170	0.435
13	3	20	160	0.364
14	3	10	150	0.469
15	3	30	170	0.392
16	4	10	160	0.444
17	4	20	150	0.45

Table A6 Analysis of variance (ANOVA) for bulk density of microencapsulated cumin oil powder

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F	Sum of Squares
Model	0.044	9	4.901E-003	19.23	0.0004	Significant
A-Carrier blend ratio	5.512E-003	1	5.512E-003	21.64	0.0023	
B- Core concentration	3.160E-003	1	3.160E-003	12.40	0.0097	
C- Inlet temperature	0.011	1	0.011	42.12	0.0003	
AB	3.610E-004	1	3.610E-004	1.42	0.2727	
AC	1.681E-003	1	1.681E-003	6.60	0.0371	
BC	4.202E-004	1	4.202E-004	1.65	0.2399	
A^2	2.679E-003	1	2.679E-003	10.52	0.0142	
B^2	0.011	1	0.011	44.64	0.0003	
C^2	6.072E-003	1	6.072E-003	23.83	0.0018	
Residual	1.784E-003	7	2.548E-004			
Lack of Fit	1.443E-003	3	4.809E-004	5.64	0.0639	Not significant
Pure Error	3.408E-004	4	8.520E-005			
Cor Total	0.046	16				
Std. Dev.	0.016		R-Squared		0.9611	
Mean	0.40		Adj R-Squared		0.9112	
C.V.%	3.94		Pred R-Squared		0.4854	
PRESS	0.024		Adeq Precision		12.773	

Table A7 Effect of process parameters on wettability of microencapsulated cumin oil powder

Run	Carrier blend ratio	Core concentration (%)	Inlet temperature (°C)	Wettability (s)
1	3	20	160	190
2	4	30	160	264
3	3	30	150	150.2
4	3	20	160	200.1
5	**4**	**20**	**170**	**275**
6	3	20	160	186.6
7	2	10	160	129.8
8	2	30	160	143.3
9	**2**	**20**	**150**	**120.3**
10	2	20	170	210.1
11	3	20	160	175.3
12	3	10	170	215.6
13	3	20	160	189.2
14	3	10	150	136.8
15	3	30	170	246.8
16	4	10	160	221.3
17	4	20	150	235.3

Table A8 Analysis of variance (ANOVA) for wettability of microencapsulated cumin oil powder

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F	Sum of Squares
Model	34490.94	9	3832.33	36.98	< 0.0001	Significant
A-Carrier blend ratio	19217.80	1	19217.80	185.45	< 0.0001	
B- Core concentration	1270.08	1	1270.08	12.26	0.0100	
C- Inlet temperature	11620.50	1	11620.50	112.14	< 0.0001	
AB	213.16	1	213.16	2.06	0.1946	
AC	627.50	1	627.50	6.06	0.0434	
BC	79.21	1	79.21	0.76	0.4110	
A2	615.70	1	615.70	5.94	0.0449	
B2	485.00	1	485.00	4.68	0.0673	
C2	407.89	1	407.89	3.94	0.0877	
Residual	725.39	7	103.63			
Lack of Fit	410.58	3	136.86	1.74	0.2970	Not significant
Pure Error	314.81	4	78.70			
Cor Total	35216.34	16				
Std. Dev.	10.18		R-Squared		0.9794	
Mean	193.51		Adj R-Squared		0.9529	
C.V.%	5.26		Pred R-Squared		0.7995	
PRESS	7061.21		Adeq Precision		22.318	

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Table A9 Effect of process parameters on cold water solubility of microencapsulated cumin oil powder

Run	Carrier blend ratio	Core concentration (%)	Inlet temperature (°C)	Cold water solubility (%)
1	3	20	160	93.89
2	4	30	160	92.53
3	3	30	150	89.09
4	3	20	160	93.75
5	4	20	170	91.09
6	3	20	160	93.87
7	2	10	160	93.85
8	**2**	**30**	**160**	**94.89**
9	2	20	150	89.99
10	2	20	170	94.95
11	3	20	160	94.75
12	3	10	170	91.22
13	3	20	160	94.08
14	3	10	150	87.63
15	3	30	170	94.15
16	4	10	160	88.67
17	**4**	**20**	**150**	**87.55**

Table A10 Analysis of variance (ANOVA) for cold water solubility of microencapsulated cumin oil powder

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F	Sum of Squares
Model	110.25	9	12.25	99.15	< 0.0001	Significant
A-Carrier blend ratio	23.94	1	23.94	193.80	< 0.0001	
B- Core concentration	10.79	1	10.79	87.32	< 0.0001	
C- Inlet temperature	36.77	1	36.77	297.59	< 0.0001	
AB	1.99	1	1.99	16.09	0.0051	
AC	0.50	1	0.50	4.08	0.0831	
BC	0.54	1	0.54	4.37	0.0748	
A2	1.54	1	1.54	12.48	0.0096	
B2	4.03	1	4.03	32.58	0.0007	
C2	27.76	1	27.76	224.71	< 0.0001	
Residual	0.86	7	0.12			
Lack of Fit	0.23	3	0.076	0.48	0.7159	Not significnt
Pure Error	0.64	4	0.16			
Cor Total	111.11	16				
Std. Dev.	0.35		R-Squared		0.9922	
Mean	92.11		Adj R-Squared		0.9822	
C.V.%	0.38		Pred R-Squared		0.9583	
PRESS	4.64		Adeq Precision		28.739	

Table A11 Effect of process parameters on colour of microencapsulated cumin oil powder

Run	Carrier blend ratio	Core concentration (%)	Inlet temperature (°C)	Colour (L*)
1	3	20	160	87.29
2	4	30	160	88.31
3	3	30	150	86.79
4	3	20	160	87.55
5	4	20	170	86.52
6	3	20	160	87.66
7	2	10	160	86.74
8	2	30	160	85.68
9	2	20	150	85.75
10	**2**	**20**	**170**	**84.59**
11	3	20	160	87.85
12	3	10	170	85.35
13	3	20	160	87.75
14	3	10	150	88.05
15	3	30	170	84.65
16	4	10	160	88.75
17	**4**	**20**	**150**	**88.92**

Table A12 Analysis of variance (ANOVA) for colour of microencapsulated cumin oil powder

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F	Sum of Squares
Model	28.89	9	3.21	50.74	< 0.0001	Significant
A-Carrier blend ratio	11.86	1	11.86	187.42	< 0.0001	
B- Core concentration	1.50	1	1.50	23.65	0.0018	
C- Inlet temperature	8.82	1	8.82	139.40	< 0.0001	
AB	0.096	1	0.096	1.52	0.2576	
AC	0.38	1	0.38	6.08	0.0432	
BC	0.078	1	0.078	1.24	0.3024	
A2	2.368E-004	1	2.368E-004	3.743E-003	0.9529	
B2	0.25	1	0.25	3.91	0.0884	
C2	5.74	1	5.74	90.71	< 0.0001	
Residual	0.44	7	0.063			
Lack of Fit	0.26	3	0.086	1.86	0.2778	Not significant
Pure Error	0.19	4	0.046			
Cor Total	29.34	16				
Std. Dev.	0.25		R-Squared		0.9849	
Mean	86.95		Adj R-Squared		0.9655	
C.V.%	0.29		Pred R-Squared		0.8496	
PRESS	4.41		Adeq Precision		23.507	

Table A13 Effect of process parameters on encapsulation efficiency of microencapsulated cumin oil powder

Run	Carrier blend ratio	Core concentration (%)	Inlet temperature (°C)	Encapsulation efficiency (%)
1	3	20	160	91.56
2	4	30	160	78.41
3	3	30	150	71.41
4	3	20	160	90.41
5	4	20	170	75.56
6	3	20	160	91.21
7	2	10	160	93.81
8	2	30	160	84.21
9	2	20	150	80.99
10	**2**	**20**	**170**	**93.88**
11	3	20	160	87.42
12	3	10	170	92.33
13	3	20	160	89.21
14	3	10	150	73.11
15	3	30	170	85.24
16	4	10	160	83.42
17	**4**	**20**	**150**	**68.56**

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Table A14 Analysis of variance (ANOVA) for encapsulation efficiency of microencapsulated cumin oil powder

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F	Sum of Squares
Model	1017.10	9	113.01	12.41	0.0016	Significant
A-Carrier blend ratio	275.42	1	275.42	30.26	0.0009	
B- Core concentration	68.44	1	68.44	7.52	0.0288	
C- Inlet temperature	350.33	1	350.33	38.49	0.0004	
AB	5.27	1	5.27	0.58	0.4717	
AC	8.67	1	8.67	0.95	0.3615	
BC	7.26	1	7.26	0.80	0.4014	
A2	35.10	1	35.10	3.86	0.0903	
B2	18.79	1	18.79	2.06	0.1940	
C2	226.06	1	226.06	24.83	0.0016	
Residual	63.72	7	9.10			
Lack of Fit	52.38	3	17.46	6.16	0.0557	Not significant
Pure Error	11.34	4	2.83			
Cor Total	1080.82	16				
Std. Dev.	3.02		R-Squared		0.9410	
Mean	84.16		Adj R-Squared		0.8652	
C.V.%	3.58		Pred R-Squared		0.2082	
PRESS	855.82		Adeq Precision		11.308	

APPENDIX B

Table B1 Effect of process parameters on moisture content of nanoencapsulated cumin oil powder

Run	Carrier blend ratio	Core concentration (%)	Inlet air temperature (°C)	Moisture content (w.b.) (%)
1	4	30	160	3.14
2	3	30	150	4.01
3	**2**	**20**	**150**	**4.23**
4	3	20	160	3.49
5	**4**	**20**	**170**	**2.7**
6	3	20	160	3.2
7	2	10	160	3.81
8	2	30	160	3.93
9	3	10	150	4.1
10	3	20	160	3.51
11	3	30	170	2.83
12	3	20	160	3.35
13	2	20	170	3.45
14	4	10	160	3.07
15	3	10	170	2.9
16	3	20	160	3.25
17	4	20	150	3.72

Table B2 Analysis of variance (ANOVA) for moisture content of nanoencapsulated cumin oil powder

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F	
Model	3.24	9	0.36	17.55	0.0005	Significant
A-Carrier blend ratio	0.97	1	0.97	47.41	0.0002	
B- Core concentration	1.125E-004	1	1.125E-004	5.481E-003	0.9431	
C- Inlet temperature	2.18	1	2.18	106.41	< 0.0001	
AB	6.250E-004	1	6.250E-004	0.030	0.8664	
AC	0.014	1	0.014	0.70	0.4299	
BC	1.000E-004	1	1.000E-004	4.872E-003	0.9463	
A^2	0.039	1	0.039	1.90	0.2105	
B^2	4.112E-003	1	4.112E-003	0.20	0.6680	
C^2	0.020	1	0.020	0.97	0.3576	
Residual	0.14	7	0.021			
Lack of Fit	0.066	3	0.022	1.15	0.4316	Not significant
Pure Error	0.077	4	0.019			
Cor Total	3.38	16				
Std. Dev.	0.14		R-Squared		0.9576	
Mean	3.45		Adj R-Squared		0.9030	
C.V.%	4.15		Pred R-Squared		0.6501	
PRESS	1.18		Adeq Precision		15.858	

Table B3 Effect of process parameters on water activity of nanoencapsulated cumin oil powder

Run	Carrier blend ratio	Core concentration (%)	Inlet air temperature (°C)	Water activity
1	4	30	160	0.236
2	3	30	150	0.315
3	**2**	**20**	**150**	**0.338**
4	3	20	160	0.256
5	**4**	**20**	**170**	**0.205**
6	3	20	160	0.241
7	2	10	160	0.301
8	2	30	160	0.295
9	3	10	150	0.322
10	3	20	160	0.225
11	3	30	170	0.218
12	3	20	160	0.261
13	2	20	170	0.27
14	4	10	160	0.225
15	3	10	170	0.214
16	3	20	160	0.239
17	4	20	150	0.285

Table B4 Analysis of variance (ANOVA) for water activity of nanoencapsulated cumin oil powder

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F	
Model	0.026	9	2.891E-003	15.84	0.0007	Significant
A-Carrier blend ratio	8.001E-003	1	8.001E-003	43.84	0.0003	
B- Core concentration	5.000E-007	1	5.000E-007	2.740E-003	0.9597	
C- Inlet temperature	0.016	1	0.016	85.35	< 0.0001	
AB	7.225E-005	1	7.225E-005	0.40	0.5492	
AC	3.600E-005	1	3.600E-005	0.20	0.6703	
BC	3.025E-005	1	3.025E-005	0.17	0.6961	
A^2	7.731E-004	1	7.731E-004	4.24	0.0786	
B^2	1.671E-004	1	1.671E-004	0.92	0.3705	
C^2	1.153E-003	1	1.153E-003	6.32	0.0402	
Residual	1.277E-003	7	1.825E-004			
Lack of Fit	4.502E-004	3	1.501E-004	0.73	0.5875	Not significant
Pure Error	8.272E-004	4	2.068E-004			
Cor Total	0.027	16				
Std. Dev.	0.014		R-Squared		0.9532	
Mean	0.26		Adj R-Squared		0.8930	
C.V.%	5.17		Pred R-Squared		0.6887	
PRESS	8.496E-003		Adeq Precision		14.622	

Table B5 Effect of process parameters on bulk density of nanoencapsulated cumin oil powder

Run	Carrier blend ratio	Core concentration (%)	Inlet air temperature (°C)	Bulk density (g/cm^3)
1	4	30	160	0.395
2	3	30	150	0.481
3	2	20	150	0.493
4	3	20	160	0.401
5	**4**	**20**	**170**	**0.37**
6	3	20	160	0.404
7	**2**	**10**	**160**	**0.495**
8	2	30	160	0.44
9	3	10	150	0.493
10	3	20	160	0.401
11	3	30	170	0.417
12	3	20	160	0.381
13	2	20	170	0.45
14	4	10	160	0.47
15	3	10	170	0.469
16	3	20	160	0.395
17	4	20	150	0.479

Table B6 Analysis of variance (ANOVA) for bulk density of nanoencapsulated cumin oil powder

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F	Sum of Squares
Model	0.029	9	3.266E-003	15.58	0.0008	Significant
A-Carrier blend ratio	3.362E-003	1	3.362E-003	16.03	0.0052	
B- Core concentration	4.705E-003	1	4.705E-003	22.44	0.0021	
C- Inlet temperature	7.200E-003	1	7.200E-003	34.34	0.0006	
AB	1.000E-004	1	1.000E-004	0.48	0.5121	
AC	1.089E-003	1	1.089E-003	5.19	0.0567	
BC	4.000E-004	1	4.000E-004	1.91	0.2097	
A^2	1.410E-003	1	1.410E-003	6.73	0.0358	
B^2	5.247E-003	1	5.247E-003	25.02	0.0016	
C^2	4.669E-003	1	4.669E-003	22.27	0.0022	
Residual	1.468E-003	7	2.097E-004			
Lack of Fit	1.129E-003	3	3.762E-004	4.44	0.0921	Not significant
Pure Error	3.392E-004	4	8.480E-005			
Cor Total	0.031	16				
Std. Dev.	0.014		R-Squared		0.9524	
Mean	0.44		Adj R-Squared		0.8913	
C.V.%	3.31		Pred R-Squared		0.3977	
PRESS	0.019		Adeq Precision		11.548	

Table B7 Effect of process parameters on wettability of nanoencapsulated cumin oil powder

Run	Carrier blend ratio	Core concentration (%)	Inlet air temperature (°C)	Wettability (s)
1	4	30	160	339.8
2	3	30	150	188.9
3	2	20	150	163.5
4	3	20	160	225.4
5	**4**	**20**	**170**	**364.8**
6	3	20	160	217.2
7	**2**	**10**	**160**	**150.2**
8	2	30	160	175.8
9	3	10	150	170.4
10	3	20	160	239.3
11	3	30	170	305.1
12	3	20	160	218.2
13	2	20	170	254.3
14	4	10	160	270.5
15	3	10	170	263.5
16	3	20	160	233.1
17	4	20	150	287.2

Table B8 Analysis of variance (ANOVA) for wettability of nanoencapsulated cumin oil powder

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F	
Model	59389.80	9	6598.87	44.50	< 0.0001	Significant
A-Carrier blend ratio	33605.28	1	33605.28	226.60	< 0.0001	
B- Core concentration	3003.13	1	3003.13	20.25	0.0028	
C- Inlet temperature	17832.16	1	17832.16	120.24	< 0.0001	
AB	477.42	1	477.42	3.22	0.1159	
AC	43.56	1	43.56	0.29	0.6047	
BC	133.40	1	133.40	0.90	0.3745	
A^2	1938.18	1	1938.18	13.07	0.0086	
B^2	827.62	1	827.62	5.58	0.0502	
C^2	1577.33	1	1577.33	10.64	0.0138	
Residual	1038.12	7	148.30			
Lack of Fit	674.23	3	224.74	2.47	0.2014	Not significant
Pure Error	363.89	4	90.97			
Cor Total	60427.92	16				
Std. Dev.	12.18		R-Squared		0.9828	
Mean	239.25		Adj R-Squared		0.9607	
C.V.%	5.09		Pred R-Squared		0.8121	
PRESS	11356.30		Adeq Precision		23.988	

Table B9 Effect of process parameters on cold water solubility of nanoencapsulated cumin oil powder

Run	Carrier blend ratio	Core concentration (%)	Inlet air temperature (°C)	Cold water solubility (%)
1	4	30	160	95.52
2	3	30	150	93.08
3	2	20	150	93.75
4	3	20	160	96.89
5	4	20	170	94.99
6	3	20	160	97.09
7	2	10	160	96.89
8	**2**	**30**	**160**	**97.77**
9	3	10	150	91.7
10	3	20	160	96.75
11	3	30	170	97.12
12	3	20	160	96.86
13	2	20	170	97.55
14	4	10	160	92.69
15	3	10	170	94.23
16	3	20	160	96.68
17	**4**	**20**	**150**	**91.45**

Table B10 Analysis of variance (ANOVA) for cold water solubility of nanoencapsulated cumin oil powder

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F	
Model	71.82	9	7.98	105.90	< 0.0001	Significant
A-Carrier blend ratio	15.99	1	15.99	212.21	< 0.0001	
B- Core concentration	7.96	1	7.96	105.64	< 0.0001	
C- Inlet temperature	24.19	1	24.19	320.99	< 0.0001	
AB	0.95	1	0.95	12.62	0.0093	
AC	0.017	1	0.017	0.22	0.6502	
BC	0.57	1	0.57	7.57	0.0285	
A^2	0.57	1	0.57	7.53	0.0288	
B^2	2.49	1	2.49	33.09	0.0007	
C^2	17.73	1	17.73	235.29	< 0.0001	
Residual	0.53	7	0.075			
Lack of Fit	0.43	3	0.14	5.83	0.0607	Not significant
Pure Error	0.098	4	0.025			
Cor Total	72.34	16				
Std. Dev.	0.27		R-Squared		0.9927	
Mean	95.35		Adj R-Squared		0.9833	
C.V.%	0.29		Pred R-Squared		0.9029	
PRESS	7.02		Adeq Precision		29.948	

Table B11 Effect of process parameters on colour of nanoencapsulated cumin oil powder

Run	Carrier blend ratio	Core concentration (%)	Inlet air temperature (°C)	Colour (L*)
1	4	30	160	90.01
2	3	30	150	89.80
3	2	20	150	88.64
4	3	20	160	90.32
5	4	20	170	89.05
6	3	20	160	90.79
7	2	10	160	89.79
8	2	30	160	88.10
9	3	10	150	90.83
10	3	20	160	90.76
11	3	30	170	87.75
12	3	20	160	90.16
13	**2**	**20**	**170**	**87.69**
14	4	10	160	91.12
15	3	10	170	88.95
16	3	20	160	90.57
17	**4**	**20**	**150**	**91.80**

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Table B12 Analysis of variance (ANOVA) for colour of nanoencapsulated cumin oil powder

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F	
Model	23.35	9	2.59	32.74	< 0.0001	Significant
A-Carrier blend ratio	7.53	1	7.53	95.00	< 0.0001	
B- Core concentration	3.16	1	3.16	39.92	0.0004	
C- Inlet temperature	7.28	1	7.28	91.85	< 0.0001	
AB	0.084	1	0.084	1.06	0.3372	
AC	0.81	1	0.81	10.22	0.0151	
BC	7.225E-003	1	7.225E-003	0.091	0.7714	
A^2	0.68	1	0.68	8.56	0.0222	
B^2	0.56	1	0.56	7.03	0.0329	
C^2	2.86	1	2.86	36.06	0.0005	
Residual	0.55	7	0.079			
Lack of Fit	0.25	3	0.084	1.11	0.4430	Not significant
Pure Error	0.30	4	0.076			
Cor Total	23.90	16				
Std. Dev.	0.28		R-Squared		0.9768	
Mean	89.77		Adj R-Squared		0.9470	
C.V.%	0.31		Pred R-Squared		0.8115	
PRESS	4.51		Adeq Precision		18.349	

Table B13 Effect of process parameters on encapsulation efficiency of nanoencapsulated cumin oil powder

Run	Carrier blend ratio	Core concentration (%)	Inlet air temperature (°C)	Encapsulation efficiency (%)
1	4	30	160	76.53
2	3	30	150	68.51
3	2	20	150	78
4	3	20	160	87.72
5	4	20	170	73.49
6	3	20	160	85.43
7	2	10	160	89.59
8	2	30	160	81.13
9	3	10	150	73.5
10	3	20	160	84.11
11	3	30	170	82.59
12	3	20	160	87.01
13	**2**	**20**	**170**	**89.62**
14	4	10	160	80.31
15	3	10	170	88.45
16	3	20	160	86.15
17	**4**	**20**	**150**	**65.35**

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Table B14 Analysis of variance (ANOVA) for encapsulation efficiency of nanoencapsulated cumin oil powder

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F	
Model	839.66	9	93.30	14.02	0.0011	Significant
A-Carrier blend ratio	227.48	1	227.48	34.17	0.0006	
B- Core concentration	66.64	1	66.64	10.01	0.0158	
C- Inlet temperature	297.56	1	297.56	44.70	0.0003	
AB	5.48	1	5.48	0.82	0.3946	
AC	3.03	1	3.03	0.45	0.5217	
BC	0.19	1	0.19	0.028	0.8709	
A^2	35.92	1	35.92	5.40	0.0532	
B^2	6.83	1	6.83	1.03	0.3450	
C^2	180.55	1	180.55	27.12	0.0012	
Residual	46.60	7	6.66			
Lack of Fit	38.73	3	12.91	6.57	0.0503	Not significant
Pure Error	7.86	4	1.97			
Cor Total	886.25	16				
Std. Dev.	2.58		R-Squared		0.9474	
Mean	81.03		Adj R-Squared		0.8798	
C.V.%	3.18		Pred R-Squared		0.2868	
PRESS	632.04		Adeq Precision		12.748	

APPENDIX C

Estimation of Cost of Production of Microencapsulated Cumin Oil Powder

Cost of machineries

Cost of spray dryer	=	Rs. 12,00,000/-
Cost of high shear emulsifier	=	Rs. 25,000/-
Cost of sealing machine	=	Rs. 1,500/-
Miscellaneous item	=	Rs. 10,000/-
Total cost	=	Rs. 1,236,500/-
Floor space 5 m^2	=	Rs. 3230/-

Assumptions

Life span (L)	=	15 years
Working hour per shift	=	8
Number of shifts per day	=	2
Annual working hours (H)	=	4800 (300 days)
Salvage value (S)	=	10% of initial cost
Interest on initial cost (i)	=	15% annually
Repair and maintenance	=	10% of initial cost
Insurance and taxes	=	2% of initial cost

Electricity charge = Rs. 7/unit

Labour wages/person = Rs. 350/day

1. Total fixed cost per year

i. Depreciation = $\frac{C-S}{L\times H}=\frac{1,236,500-123,650}{15\times 4800}$

= Rs. 15.46/h

= Rs. 74208/year

ii. Interest = $\frac{C+S}{2}\times\frac{i}{H}=\frac{1,236,500+123,650}{2}\times\frac{15}{100\times 4800}$

= Rs. 21.25/h

= Rs. 102000/year

iii. Insurance and taxes = 2% of initial cost

$$\frac{2}{100\times 4800}\times 1,236,500$$

= Rs. 5.15/h

= Rs. 24720/year

Total fixed cost/year = (I + ii + iii)

= Rs.200928/-

2. Total variable cost per year

i. Repair& maintenance

= 10% of initial cost

$$\frac{10}{100\times 4800}\times 1{,}236{,}500$$

= Rs. 25.76/h

= Rs. 123,648/year

ii. Electricity cost

a) Energy consumed by the spray dryer = 5.8 kwh

b) Energy consumed by mixer, sealing machine and weighing balance = 0.2 kwh

Total energy consumption = 6.0 kwh

Cost of energy consumption per year = Power × Duration × Cost of 1 unit

= 6×4800×7= Rs. 201,600/-

iii. Labour charge (1 person) per shift = Rs. 350/-

Labour charge per day = 700/-

Cost of labour per year = Rs. 210,000/-

iv. Packaging cost = Rs. 50/day

= Rs. 15,000/year

v. Cost of raw materials for microencapsulated cumin oil

Quantity of cumin oil required = Rs. 1314 kg/year

Cost of cumin oil = Rs. 2950/kg

= Rs. 336,300/year

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Quantity of gum arabic required	=	291 kg/year
Cost of gum arabic	=	Rs. 1000/kg
	=	Rs. 291,000/year
Quantity of maltodextrin required	=	765 kg/year
Cost of maltodextrin	=	Rs. 90/kg
	=	Rs. 68,850/year
Cost of major raw materials/year	=	Rs. 696,150/-
Total variable cost for microencapsulated cumin oil/year	=	i+ii+iii+iv+v
	=	Rs. 1,246,398/-
Total cost for production of microencapsulated cumin oil/year	=	(Fixed cost+Variable cost)×
	=	200928+ 1,246,398
	=	Rs. 1447326/-
Total cost for production of 1 kg of microencapsulated cumin oil	=	Rs. 1577/kg

The market selling price of 1 kg of microencapsulated cumin oil= Rs. 5000/kg

$$\textbf{Benefit – cost ratio=} \frac{5000}{1577} = 3.17$$

Estimation of Cost of Production of Nanoencapsulated Cumin Oil Powder

Cost of machineries

Cost of spray dryer	=	Rs. 14,00,000/-
Cost of high shear emulsifier	=	Rs. 25,000/-
Cost of sealing machine	=	Rs. 1,500/-
Miscellaneous item	=	Rs. 10,000/-
Total cost	=	Rs. 1,436,500/-
Floor space 5 m^2	=	Rs. 3230/-

Assumptions

Life span (L)	=	15 years
Working hour per shift	=	8
Number of shifts per day	=	2
Annual working hours (H)	=	4800 (300 days)
Salvage value (S)	=	10% of initial cost
Interest on initial cost (i)	=	15% annually
Repair and maintenance	=	10% of initial cost
Insurance and taxes	=	2% of initial cost
Electricity charge	=	Rs. 7/unit

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Labour wages/person = Rs. 350/day

1. Total fixed cost per year

i. Depreciation = $\frac{C-S}{L\times H}=\frac{1,436,500-143,650}{15\times 4800}$

= Rs. 17.96/h

= Rs. 86208/year

ii. Interest = $\frac{C+S}{2}\times\frac{i}{H}=\frac{1,436,500+143,650}{2}\times\frac{15}{100\times 4800}$

= Rs. 24.69/h

= Rs. 118512/year

iii. Insurance and taxes = 2% of initial cost

$$\frac{2}{100\times 4800}\times 1,436,500$$

= Rs. 5.99/h

= Rs. 28752/year

Total fixed cost/year = (I + ii + iii)

= Rs.233472/-

2. Total variable cost per year

i. Repair& maintenance 10% of initial cost

= $\frac{10}{100\times 4800}\times 1,436,500$

	=	Rs. 29.93/h
	=	Rs. 143,664/year

ii. Electricity cost

a) Energy consumed by the spray dryer	=	5.8 kwh
b) Energy consumed by mixer, sealing machine and weighing balance	=	0.2 kwh
Total energy consumption	=	6.0 kwh
Cost of energy consumption per year	=	Power × Duration × Cost of 1 unit
	=	6×4800×7= Rs. 201,600/-
iii. Labour charge (1 person) per shift	=	Rs. 350/-
Labour charge per day	=	700/-
Cost of labour per year	=	Rs. 210,000/-
iv. Packaging cost	=	Rs. 6/day
	=	Rs.3600 /year

v. Cost of raw materials for nanoencapsulated cumin oil

Quantity of cumin oil required	=	Rs. 7.8 kg/year
Cost of cumin oil	=	Rs. 2950/kg
	=	Rs. 23010/year
Quantity of gum arabic required	=	19.5 kg/year

Cost of gum arabic	=	Rs. 1000/kg
	=	Rs. 19,500/year
Quantity of maltodextrin required	=	55.5 kg/year
Cost of maltodextrin	=	Rs. 90/kg
	=	Rs. 4995/year
Cost of major raw materials/year	=	Rs. 47505/-
Total variable cost for nanoencapsulated cumin oil/year	=	i+ii+iii+iv+v
	=	Rs. 606,369/-
Total cost for production of nanoencapsulated cumin oil/year	=	Fixed cost+Variablecost+Cost of floor space
	=	233,472+606,369+3230
	=	Rs. 843,071/-
Total cost for production of 1 kg of nanoencapsulated cumin oil	=	Rs. 13510.7/kg

The market selling price of 1 kg of nanoencapsulated cumin oil= Rs. 15000/kg

$$\textbf{Benefit – cost ratio=} \frac{15000}{13510.7} = 1.11$$

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RESPONSE SURFACE OPTIMISATION OF PROCESS VARIABLES FOR ENCAPSULATION OF CUMIN OIL BY SPRAY DRYING

By

SHAHAMA K

(2016-18-003)

ABSTRACT OF THE THESIS

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Kerala Agricultural University

DEPARTMENT OF PROCESSING AND FOOD ENGINEERING

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ABSTRACT

Spices are the main flavouring agents in food. Cumin (Cuminum cyminum Linn.) is one of the important commercial seed spices which belong to the umbellifereae family. Cumin is valued for its aroma, medicinal and therapeutic properties. The most important chemical component of cumin seed is essential oil content, ranging from 2.5 to 4.5%. The biological activity of the oil is lost due to the volatilisation or degradation of active compounds owing to direct exposure to heat, humidity, light, or oxygen. Encapsulation is the most suitable method which will protect the essential oil and flavour ingredients from the liquid form to solid form by coating agents. Microencapsulation and nanoencapsulation are the two encapsulating techniques commonly used. Spray drying is the most commonly used technique for encapsulation in the food industry as it is a rapid, continuous, cost-effective, reproducible and scalable process for the production of dry powders from a fluid material. The microencapsulation of cumin oil were carried out with a tall type spray dryer with twin fluid atomiser whereas the nanoencapsulation was done in a laboratory spray dryer with ultrasonic atomiser. The wall materials selected for encapsulation were gum arabic and maltodextrin. The process variables used in the study were gum arabic: maltodextrin ratio (1:2, 1:3 and 1:4), core concentration (10, 20, and 30%) and spray dryer inlet temperature (150, 160 and 170°C). The pysico-chemical properties of cumin oil and wall materials were determined. The optimisation of the encapsulation process was done with RSM (Response Surface Methodology) from the quality characteristics of the encapsulated powders. The optimised condition in microencapsulation were 1: 2.77 carrier blend ratio (gum arabic:maltodextrin), 10% core concentarion and 162.50°C spray dryer inlet temperature and that for nanoencapsulation were a carrier blend ratio of 1:2.92, core concentration of 10% and a spray dryer inlet temperature of 163.38°C. The total cost for the production of 1 kg of microencapsulted cuminn oil were Rs. 1577/kg and that for nanoencapsyulation were Rs. 13510.7/kg.