CHAPTER II

REVIEW OF LITERATURE

This chapter represents a review of the research work done by earlier researchers on garlic related to its active compounds, medicinal properties, biological properties and hurdles in value addition. The different microencapsulation techniques to overcome the hurdles in value addition and influencing parameters of emulsion and drying parameters to produce effective encapsulant are reviewed. In addition to this, the effects of process parameters on various quality parameters of the microencapsulated powder are also discussed.

2.1 GARLIC

Garlic (*Allium sativum* L.) is a bulbous perennial plant of the lily family (*Liliaceae*). A classic ingredient in many national cuisines, garlic has a powerful, onion like, aroma and pungent taste. It has been widely recognized as a valuable spice and a popular remedy for various ailments and physiological disorders. The name garlic may have originated from the Celtic word 'all' meaning pungent. It is cultivated practically throughout the world. It appears to have originated in central Asia and then spread to China, the Near East, and the Mediterranean region before moving west to Central and Southern Europe, Northern Africa (Egypt) and Mexico [33].

Fresh garlic contains water, carbohydrates, proteins, fiber and fat as well as 33 sulfur compounds, 17 amino acids, germanium (14 µg/100 g), calcium (50–90 µg/100 g), copper (0.02–0.03 µg/100 g), iron (2.8–3.9 µg/100 g), potassium (100–120 µg/100 g), magnesium (43–77 µg/100 g), chromium (0.3–0.5 mg/100 g), manganese (0.2–0.6 mg/100 g), boron (0.3–0.6 mg/100 g), barium (0.2–1 mg/100 g), aluminum (0.5–1 mg/100 g), sodium (10–22 mg/100 g), phosphorous (390–460 mg/100 g), zinc (1.8–3.1 mg/100 g), selenium (15–35 µg/100 g), thiamine (0.25 mg/100 g), riboflavin (0.08
Garlic or garlic extracts have relatively consistent effects on cancer prevention [35]. It is a common food ingredient, readily available, safer and cheaper than tablets of purified organosulfur compounds.

According to research and analysis, Allium vegetables (scallion, garlic, onion, leek) contain substances such as allicin, diallyl disulphide, and trace amounts of selenium and quercetin, of which allicin has been found to be able to kill the “superbug” MRSA (Methicillin-resistant Staphylococcus aureus) [36].

Many recent studies have provided strong evidence that most of the biological functions of garlic are attributed to allicin [1,5,10,35,36]. In fact, no compounds outside the thiosulfinates (of which allicin is about 60-80%) has been found that accounts for a significant portion of the pharmacological activities of crushed garlic at levels representing normal human consumption (2 to 5 g/day). Typical whole fresh garlic weighing 2 to 4 gram may contain from 6 to 14 mg of alliin per gram and these biological effects of thiosulfinates can be related to their strong SH -modifying and antioxidant properties [37].

In Japan and western countries, garlic products have been popular and marketed in recent years as healthy foods with beneficial physiological effects for humans. Consequently, the majority of the garlic supplements sold today is garlic powder tablets that are standardized on allicin [7, 8].

2.2 ACTIVE COMPOUNDS

The main active element in garlic is allicin, which is currently thought to improve immunity, prevent cancer and cardiovascular diseases, and function as an antibiotic. In recent years, there have been continuous research and reports into the
physical and medicinal effects of allicin. Out of the different parts of garlic, the greatest concentration of allicin is the bulb, then the leaves, and finally the root. Different organs and different parts in a same growth stage contain different allicin content due to differences in activity of alliin and alliinase [38].

Allicin in its pure form was found to exhibit i) antibacterial activity against a wide range of Gram-negative and Gram-positive bacteria, including multidrug-resistant enterotoxigenic strains of *Escherichia coli*; ii) antifungal activity, particularly against *Candida albicans*; iii) antiparasitic activity, including some major human intestinal protozoan parasites such as *Entamoeba histolytica* and *Giardia lamblia*; and iv) antiviral activity [39].

Allicin is a light yellow oily substance with a garlic smell. It has specific gravity of 1.112 (20°C), Refractive index of 1.561, no optical rotation, slightly soluble in water, soluble in organic solvents such as ethanol, benzene, diethyl ether, and other organic solvents, unstable in heat, more stable with regard to acid, irritating to the skin and diallyl thiosulfinate compound, of which the chemical structure is shown in Fig. 2.1, is
the main active ingredient in Allium vegetables [40]. For example, when garlic is mashed, the enzyme allinase present in garlic will be released, converting alliin in garlic into Allicin [41], as shown in Fig. 2.2.

2. 3 MEDICINAL PROPERTIES

Garlic possesses many healthful properties that are related to its bioactive compounds [42]. It was reported that consumption of garlic is very helpful in regulating plasma lipid levels [43] as well as plasma anticoagulant activity [44] and in prevention of the atherosclerosis process [45] and even cancer [46]. It was shown that garlic also provides protection against ethanol induced gastric injury [47]. The most studied and reported health-promoting effect of garlic is cardioprotection [45].

Hippocrates, the Father of Medicine, observed that garlic was excellent for curing tumors and is an effective diuretic. Aristotle attributed garlic as a cure for rabies, and the Prophet Mohammad recommended it for treating scorpion stings. Desired medicinal results of garlic are obtained when bulbs are chewed and swallowed or mixed with food and eaten [48].

Preparations of garlic are available as tablets, capsules, syrup, tinctures and oil. In ointment form, garlic has been used externally for treatment of ring worm; boiled with vinegar and sugar for treatment of asthma; made into an infusion for treatment of epilepsy; pounded with honey for use against rheumatism; and mixed with milk for use as a vermifuge. Garlic is commonly used in Europe and Asia for medicinal benefits in healing wounds, etc. [49]. In Germany, sale of garlic preparations competes with sales of leading drugs [48].

Successful clinical use of garlic for treating elevated blood pressure and arteriosclerosis has been known since the early part of this century. It has been reported that regular garlic intake causes both a prolonged lowering of hypertension and an improved sense of well-being in patients. As early as 1928, definite blood pressure
decreases were achieved as well as increases in productive heart power with garlic therapy, not only in older patients, but also in younger hypertonic patients [50].

The reduction of malignant disease incidence by garlic or some organosulfur compounds from garlic was reported from many countries, such as Italy and Germany by Milner [51], China [52], England [53], and US [54], which indicates cancer preventive effect of garlic is not localized to races or regions. The organosulfur compounds present in garlic, such as diallyl sulfide (DAS), diallyl disulfide (DADS), or S-allyl cysteine (SAC) have been reported as potent chemopreventive agents in stomach [55], breast [56], colon [57] and other cancers.

Garlic is used traditionally as a complementary therapy in the treatment of several diseases such as diabetes, several forms of cancer and neurodegenerative conditions such as ischemic stroke [58]. In addition, garlic has been reported to possess a range of cardiovascular effects such as lowering of plasma cholesterol; inhibition of platelet aggregation as well as reducing of arterial blood pressure [59].

The effect of garlic extract on colon cancer inhibition was examined in rats, and reported that the optimal level of garlic extract supplementation might be 2.5% of the diet (approximately 0.6 g/kg body weight) [60]. Stomach cancer incidence was decreased in the subjects consuming 20 g/day of garlic compared to the subjects consuming 1 g/day [53]. In reviews by Milner [51], 5 g/day of fresh crushed garlic were suggested to reduce urinary N-nitrosoproline excretion in human. On the contrary, some reports have shown that the high dose of garlic (more than 0.5 g/kg body weight) by gastric intubation altered the morphology of rat liver and kidney, or decreased antioxidative system, such as catalase and superoxide dismutase [61]. Therefore, the practical approach for garlic supplementation should be needed in order to know the effect of garlic on human health and chemoprevention.

Allicin which is one of the most biologically active compounds of garlic inhibited the growth of cancer cells of murine and human origin [62]. It induced the
formation of apoptotic bodies, nuclear condensation and a typical DNA ladder in cancer cells. Furthermore, activation of caspases -3, -8 and -9 and cleavage of poly (ADP-ribose) polymerase were induced by allicin.

It is also well established that garlic extracts, in particular in the form of powders can show a significant anti-cholesterol activity. A 12 week study comparing the effect of standardised garlic powder tablets (900 mg daily) with that of bezafibrate (600 mg daily), one of the most commonly prescribed blood lipid-lowering drugs until the advent of the statins, has also been conducted. The multi-centre, double-blind study was performed with 94 patients having cholesterol and/or triglyceride values exceeding 250 mg/dL. After 4 weeks of treatment, the decreases in cholesterol, LDL cholesterol, and triglyceride levels were all statistically highly significant, and there were no differences between the effects of garlic and bezafibrate. HDL cholesterol values in the course of 4 weeks also increased significantly, again without any differences between the two regimes [50].

There is a growing body of evidence that garlic may have significant enhancing effects on the immune system. While most of the work has been conducted on animals or in vitro, the human studies that have been conducted are encouraging. Preliminary studies in humans, using an alliin standardised garlic powder preparation, have demonstrated positive effects on immunoreactions and phagocytosis. In geriatric subjects, the administration of 600 mg garlic powder per day for 3 months induced significant (p<0.01) increases in the percentage of phagocytosing peripheral granulocytes and monocytes when tested ex-vivo for their ability to engulf Escherichia coli bacteria. The cell counts of lymphocyte cell sub-populations were also increased. Another human study was conducted with an unrefined garlic extract (5-10 g/day) which was given to AIDS patients. For the seven patients who completed the 12-week study, there was a major increase in the percent natural killer cell activity from a seriously low mean value of 5±4% to a more normal mean value of 36±15% [63].
2. 4 BIOLOGICAL PROPERTIES

The antimicrobial properties of garlic were first described by Pasteur in 1958, and since then, research had demonstrated its effectiveness against bacteria, protozoa, fungi and some viruses [64]. Research works have been conducted on the antimicrobial effects and drying of garlic by [65 –69]. It also indicated that garlic has anti-neoplastic, cardiovascular, immuno-stimulatory and hypoglycemic properties [70].

Fresh garlic extracts in which allicin is known to be the main active component have been shown to have in-vitro and in-vivo antiviral activity. Among the viruses which are sensitive to garlic extracts are the human cytomegalovirus, influenza B, herpes simplex virus type 1, herpes simplex virus type 2, parainfluenza virus type 3, vaccinia virus, vesicular stomatitis virus, and human rhinovirus type 2 [71].

Garlic extracts also have a strong antifungal effect and inhibit the formation of mycotoxins like the aflatoxin of Aspergillus parasiticus [72].

The antibacterial properties of crushed garlic have been known for a long time. Various garlic preparations have been shown to exhibit a wide spectrum of antibacterial activity against Gram-negative and Gram-positive bacteria including species of Escherichia, Salmonella, Staphylococcus, Streptococcus, Klebsiella, Proteus, Bacillus, and Clostridium. Even acid-fast bacteria such as Mycobacterium tuberculosis are sensitive to garlic [73]. Garlic extracts are also effective against Helicobacter pylori the cause of gastric ulcers [74]. Garlic extracts can also prevent the formation of Staphylococcus enterotoxins A, B, and C1 and also thermonuclease [75]. Cavalito and Bailey, [76] were the first to demonstrate that the antibacterial action of garlic is mainly due to Allicin. The sensitivity of various bacterial and clinical isolates to pure preparations of allicin [50] is very significant. The antibacterial effect of allicin is of a broad spectrum. In most cases the 50% lethal dose concentrations were somewhat higher than those required for some of the newer antibiotics. Interestingly, various bacterial strains resistant to antibiotics such as methicillin resistant staphylococcus
*aureus* as well as other multidrug-resistant enterotoxicogenic strains of *Escherichia coli*, *Enterococcus*, *Shigella dysenteriae*, *S. flexneni*, and *S. sonnei* cells were all found to be sensitive to Allicin.

Cavallito and Bailey [76] had demonstrated that garlic juice diluted to one part in 125,000 parts inhibits the bacterial growth of *Staphylococcus*, *Streptococcus*, *Vibrio* (including *V. cholerae*) and *Bacillus* (including *Bacillus typhosus*, *B. dysenteriae* and *B. enteritidis*). Johnson and Vaughn [77] reported that 10% extract of dehydrated garlic bulb demonstrated antibacterial action against *Salmonella typhimurium* and *Escherichia coli* within 2 to 6 h exposure. Bacterial cultures resistant to the commonly used antibiotic chloramphenicol appear sensitive to garlic.

Antioxidant capacity measured by the ferric-reducing/antioxidant power (FRAP) method and 1,1-diphenyl-2-picrylhydrazyl (DPPH) free radical assay was the highest in raw and in a short time processed garlic samples by cooking [78].

### 2.5 Hurdles in Value Addition

It has also been found that if garlic powder or granules were stored for longer periods, active ingredients present in freshly ground garlic were often eliminated or otherwise rendered inactive. The alliinase is irreversibly deactivated at the pH level in the human stomach. If taking garlic powder directly, there would be only an insignificant amount of allicin that would be effective inside the human body. Hence Li [79] conducted studies on microencapsulation of garlic powder using vacuum drying, which could resist human stomach conditions in order to prolong the shelf life and protect alliinase activity through the stomach transit.

The spice extracts are used either in the form of liquid concentrate or adsorbed on a carrier. In both cases oleoresin undergoes oxidative degradation. It was found that destruction of several pigments occur under exposure to oxygen: hydroxyl groups are converted into unstable ketones, which are then decomposed into colourless
compounds with a short carbon skeleton [12]. Apart from the fading effect, the oxidation products and intermediates (such as peroxides) may have a detrimental effect on the foodstuffs themselves. Hence, there is a need for protection of the oleoresin against environmental factors, which contribute to its deterioration, e.g. oxygen, light, moisture [11].

Although the spice oleoresins provide a more complete flavour profile than their respective essential oils, their sensitivity to light, heat and oxygen is a disadvantage. This can be overcome by effective encapsulation [80].

2.6 MICROENCAPSULATION TECHNOLOGY

Microencapsulation technology may be defined as a process to entrap one substance within another substance [15], thereby producing particles with diameters of a few nm to a few mm [81], that may offer protection against oxygen, heat, humidity and light. The microcapsules have multitudes of shapes, depending on the materials and methods used to prepare them [82] that release their contents at controlled rates over prolonged periods under specific conditions [83].

Encapsulation of lipids is a physical means to offer protection against oxidation without the need of antioxidants [84]. The process of microencapsulation not only protects against losses and chemical changes during food production and storage, but also enables production of food ingredients in the form of powders/products with new properties [85].

The purpose of encapsulation is to provide improved stability to active ingredients in final products and during processing; to minimize the degradation of volatile actives, such as aroma; to mask off-taste during eating, such as bitter taste and astringency of polyphenols; to prevent reaction with other components in food products such as oxygen or water; to immobilize cells or enzymes in food processing applications, such as fermentation process and metabolite production processes [86].
Encapsulation technology is now well developed and accepted within the pharmaceutical, chemical, cosmetic, foods and printing industries [16]. In food products, fats and oils, aroma compounds and oleoresins, vitamins, minerals, colorants, enzymes [13, 87] and microorganisms [18] have been encapsulated.

Microencapsulated fish oil and non encapsulated fish oil with the addition of antioxidants was compared by Baik et al.,[88]. This study showed that encapsulated fish oil was 10 times more stable against oxidation than non encapsulated fish oil.

2.7 TECHNIQUES OF ENCAPSULATION

2.7.1 Extrusion

Extrusion, as applied to flavour encapsulation, is a relatively low temperature entrapping method, which involves forcing a core material in a molten carbohydrate mass through a series of dies into a bath of dehydrating liquid. The pressure and temperature employed are typically <689 kPa and < 115°C. The coating material hardens on contacting the liquids, forming an encapsulating matrix to entrap the core material. The extruded filaments are then separated from the liquid bath, dried, and sized [89].

The carrier used may be composed of more than one ingredient, such as sucrose, maltodextrin, glucose syrup, glycerine and glucose [89].

The dripping tool can be simply a pipette, a syringe, a vibrating nozzle, a spraying nozzle, jet cutter or atomizing disk [81]. In comparison to other extrusion techniques, Jet cutter was found to be the best technology for large-scale/industrial applications [91].

Extrusion is commonly employed for the microencapsulation of microbial cells [92]. In employing extrusion, a polymeric solution is first mixed with the microbial cells and then extruded through an orifice as droplets into the solution of a cross-linking
agent. Gelation occurs by contact of the polymer solution with the cross-linking agent, cooling or a combination of both. The factors affecting the size of the microspheres produced include the diameter of the orifice, the viscosity and flow rate of the polymeric solution, the drop height or distance from the orifice to the cross-linking solution and concentration and temperature of the polymer solution [93].

Extrusion is used to encapsulate minerals and vitamins in oil (saturated fat) in a glassy matrix (polysaccharides), with further grinding if a powder form is required [94].

The advantage of this method is that the material is completely surrounded by the wall material (true encapsulation), and any residual oil or core material is removed from the surface in an alcohol bath. It provides an excellent stability against oxidation and therefore prolongs the shelf life. The product can be kept for 1–2 years without any substantial quality degradation [83].

The major advantages of the extrusion method are simplicity of its operation, lower cost, and mild operational conditions ensuring high cell viability [95]. However, there are also a few drawbacks such as its inefficiency in producing microspheres smaller than 500 µm, requirement of low to moderate viscosity polymer solutions and relatively large diameter nozzles [96]. Yuliani et al., [97] reported that this encapsulation technique was considered unsuitable for subsequent extrusion processing because the water in the extruder melt could dissolve the capsules.

Electrostatic extrusion is especially effective for production of very small particles, down to 50 µm. The basis of electrostatic droplet generation is the acceleration of the normal droplet formation process using electrostatic forces to pull the droplets off the orifice (needle) at a considerably faster rate compared to the normal dripping process, whereby removal is based solely on gravitational force. The electrical potential, which can be static or pulsed is applied to the extruded polymer solution by passing it through a charged needle, with the produced droplets subsequently falling into a collecting/hardening solution, which has being earthed or holds an opposite
charge. If the electrodes are parallel plates, a uniform electric field is generated with respect to direction and strength, thus a uniform force is applied to the droplets at the tip of the nozzle. It has been reported that the strong electric fields do not cause cells to lose viability and activity during the encapsulation process. This technique is capable of producing smaller microbeads compared to normal dripping of uniform size and shape under reproducible conditions and can also be performed under sterile conditions. The main disadvantages include low production rates due to the low polymer flow rate through the needle, although this can be improved by increasing the number of needles. An alternative extrusion technology is co-extrusion. It might be utilized to prepare spherical microbeads with a hydrophobic core and a hydrophilic or hydrophobic shell [98].

2.7.2 Co-extrusion / Centrifugal Extrusion

A number of food-approved coating systems have been formulated to encapsulate products such as flavourings, seasonings, and vitamins. These wall materials include gelatin, sodium alginate, carrageenan, starches, cellulose derivatives, gum acacia, fats, fatty acids, waxes, and polyethylene glycol [99].

Centrifugal extrusion is a liquid co-extrusion process utilizing nozzles consisting of concentric orifice located on the outer circumference of a rotating cylinder (i.e., head). The encapsulating cylinder or head consists of a concentric feed tube through which coating and core materials are pumped separately to the many nozzles mounted on the outer surface of the device. While the core material passes through the center tube, coating material flows through the outer tube. The entire device is attached to a rotating shaft such that the head rotates around its vertical axis. As the head rotates, the core and coating materials are co-extruded through the concentric orifices of the nozzles as a fluid rod of the core sheathed in coating material. Centrifugal force impels the rod outward, causing it to break into tiny particles. By the action of surface tension, the coating material envelops the core material, thus accomplishing encapsulation. The
Microcapsules are collected on a moving bed of fine-grained starch, which cushions their impact and absorbs unwanted coating moisture. Particles produced by this method have diameter ranging from 150 to 2000 µm [99].

### 2.7.3 Air Suspension Coating / Fluidized Bed Coating

Air-suspension particle coating is a process where thin coatings are applied to powder particles in a batch processor or a continuous set-up. It is different than spray drying encapsulation, which produces particles consisting of a homogeneously blended matrix of the polymer entrapping the particle [87]. The basic principle of both air-suspension particle coating and air-suspension agglomeration is to atomize a fine liquid spray into a bed of fluidized particles. The spray consists of a solute which acts as a coating medium, and a solvent in which the solute is dissolved or slurried. The liquid impinges and spreads on the particles. The fluidization air evaporates the solvent, leaving a layer of solute on the surface of the particle. Particle growth can occur by either inter-particle agglomeration or surface layering. The different fluidized-bed coating methods are: top-spray, bottom-spray, and tangential spray [82].

The coating can be selected from cellulose derivatives, dextrins, emulsifiers, lipids, protein derivatives and starch derivatives [13].

The two most important processing variables in this process are volume of fluidized air used, which controls the height of the substrate particles in the air stream and determines their surface coating time. The other variable is air temperature. This is a critical factor as improper temperature control will result in incomplete coverage by the coating material and thus lead to a poor quality product. This method is common for use in the nutritional supplement market to supply encapsulated versions of vitamin C, vitamin B, ferrous sulphate, ferrous fumarate, sodium ascorbate, potassium chloride and a variety of vitamin/mineral premixes. It has applications in baked goods, seasonings, fillings, desserts and dry mix puddings [100].
2.7.4 Spray Chilling / Spray Cooling

Spray-chilling or Spray-cooling are technologies to produce lipid-coated active agents. With the ability to select the melting point of the wall, these methods of encapsulation can be used for controlled release [100]. In spray-chilling and spray-cooling, the core and wall mixtures are atomized into the cooled or chilled air, which causes the wall to solidify around the core. Unlike spray-drying, spray-chilling or spray-cooling does not involve evaporation of water and therefore there is no mass transfer [101]. In spray chilling, the coating is typically a fractionated or hydrogenated vegetable oil with a melting point in the range of 32 – 42°C. In spray cooling, the wall is typically a vegetable oil, although other materials can be used. The normal melting point is 45 - 122°C. These two methods which differ only in the melting point of the wall material used [100]. The microcapsules prepared by spray-chilling and spray cooling are insoluble in water due to the lipid coating [101].

Frozen liquids, heat-sensitive materials and those not soluble in the usual solvents can be encapsulated by spray chilling / spray cooling. It is routinely used for the encapsulation of a number of organic and inorganic salts like ferrous sulfate, vitamin, mineral or acidulents as well as textural ingredients, enzymes, flavors and other functional ingredients to improve heat stability, delay release in wet environments, and/or convert liquid hydrophilic ingredient into free flowing powders. [102].

The spray chilling method was devised to overcome the problem of cell damage due to the exposure of microbial cells to high temperature during spray drying. The process is performed using equipment similar to that used for spray drying except that a cold conveying air or cold chamber is used instead of hot air. Pedroso et al., [103] encapsulated B. lactis and Lactobacillus acidophilus in solid lipid microcapsules using spray chilling technique. An emulsion containing probiotic cells, molten fat and lecithin was prepared by homogenization. This emulsion was then atomized into a cold chamber.
where the solid lipid microspheres were formed. The conditions used in the spray chilling method were sufficiently mild and did not affect the viability of the encapsulated cells.

2.7.5 Lyophilization / Freeze-Drying

It is a process used for the dehydration of most heat-sensitive materials and aromas. It has been used to encapsulate water-soluble essences and natural aromas as well as drugs. Except for the long dehydration period required (commonly 20 h), freeze-drying is a simple technique, which is particularly suitable for the encapsulation of aromatic materials. The retention of volatile compounds during the lyophilization is dependent upon the chemical nature of the system [104].

Heinzelmann et al., [105] found that the production of dried microencapsulated fish oil by freezing and subsequent freeze-drying offers an opportunity to achieve a product with good oxidation stability.

Freeze drying has abundant advantages that can extend the shelf-life of protein, liposome, and nanocapsules; minimize physicochemical dissociations to maintain heat sensitive biological materials; entrap aroma compounds into defined porous structures for coffee and tea in the food industry [106 – 109]; and reduce weight for easy handling or transportation [110].

Freeze drying has been considered as a good technique to improve the long-term stability of colloidal nanoparticles [110]. It is also considered as advantageous because it maintains a higher stability of active ingredients than other drying methods [111].

2.7.6 Coacervation / Phase Separation

Coacervation involves the separation of a liquid phase of coating material from a polymeric solution followed by the coating of that phase as a uniform layer around
suspended core particles. The coating is then solidified. In general, the batch-type coacervation process consists of three steps and is carried out under continuous agitation, (i). formation of a three-immiscible chemical phase, (ii). deposition of the coating and (iii). solidification of the coating [102].

Both, simple and complex methods of coacervation can be used. In simple coacervation, a nonsolvent or a more water-soluble polymer is used. The polymer competes for the solubility for gelatin protein solution by hydrophobic interaction. In complex coacervation, the capsule is formed by the ionic interaction of two oppositely charged polymers, commonly the positive charges on protein molecules and anionic macromolecules such as gelatin and gum arabic [113]. The complex coacervate is produced when the two opposite charges are neutralized with each other [114].

The more important processing factors to be considered for the coacervation technique are the volume of the dispersed phase, addition rate of the incompatible polymer to the coating polymer solution, stirring rate of the dispersion and core material to be encapsulated. Apart from these factors, the composition and viscosity of the coacervate and supernatant phases are known to affect the size distribution, surface morphology and internal porosity of the final microspheres [115].

Microencapsulation using the coacervation technique has been attempted to encapsulate flavour oils, preservatives, enzymes as well as microbial cells [116].

A large numbers of coating materials have been evaluated for coacervation microencapsulation but the most studied and well understood coating system is gelatin/gum acacia system. However, other coating systems such as gliadin, heparin/gelatin, carrageenan, chitosan, soy protein, polyvinyl alcohol, gelatin / carboxy methyl cellulose, β-lactoglobulin/gum acacia, and guar gum/dextran are also suitable for coacervation microencapsulation. In recent years, modified coacervation processes have also been developed that can overcome some of the problems encountered during a
typical gelatin/gum acacia complex coacervation process, especially when dealing with encapsulation of heat-sensitive food ingredients such as volatile flavor oils [102].

2.7.7 Liposomes

Liposomes are single or multi-layered vesicles, which involve the complete enclosure of an aqueous phase within a phospholipid-based membrane. These vesicles form spontaneously when phospholipids are dispersed in an aqueous media [100]. The phospholipids make up the outer layer of the liposomes. The hydrophilic portions are positioned towards the aqueous phase and the hydrophobic groups associate with the hydrophobic portions of other lipid molecules. It forms a sheet which when folded into a spherical shape, results in a very stable capsule. Aqueous or lipid-soluble materials, but not both, are entrapped in these membranes and this forms the basis of encapsulation. Flavour compounds are commonly encapsulated using this method. Kirby et al., [117] used this material to encapsulate ascorbic acid with high efficiency and a very stable material. Liposomes can be made in sizes ranging from a few nanometers to several microns [100]. The most stable liposomes are made from lecithin, cholesterol and negatively charged phospholipid.

2.7.8 Molecular Inclusion

Molecular inclusion in cyclodextrins provides some specific features to bioactives; however these techniques are more expensive, and therefore, less exploited. β-cyclodextrin is a cyclic derivative of starch made up of seven glucopyranose units. The external part of the cyclodextrin molecule is hydrophilic, whereas the internal part is hydrophobic. The guest molecules, which are apolar, can be entrapped into the apolar internal cavity through a hydrophobic interaction [118]. In this method, the flavour compounds are entrapped inside the hollow centre of a β-cyclodextrin molecule [87].
2.7.9 Emulsification

The emulsification technique is widely used for encapsulation of various microbial cells [119]. It involves dispersion of the cell/polymer suspension (dispersed phase) in an oil/organic phase (continuous phase). The mixture is homogenized to form a water-in-oil emulsion with the aid of surfactant and stirring. Coagulation of the dispersed phase is initiated by cooling or addition of a cross-linking agent to the emulsion. The microspheres produced are subsequently harvested by filtration or centrifugation.

2.7.10 Cocrystallization

Cocrystallization is a new encapsulation process utilizing sucrose as a matrix for the incorporation of core materials. The sucrose syrup is concentrated to the supersaturated state and maintained at a temperature high enough to prevent crystallization. A predetermined amount of core material is then added to the concentrated syrup with vigorous mechanical agitation, thus providing nucleation for the sucrose ingredient mixture to crystallize. As the syrup reaches the temperature at which transformation and crystallization begin, a substantial amount of heat is emitted. Agitation is continued in order to promote and extend transformation crystallization until the agglomerates are discharged from the vessel. The encapsulated products are then dried to the desired moisture (if necessary) and screened to a uniform size. It is very important to properly control the rates of nucleation and crystallization as well as the thermal balance during the various phases [120].

The advantages of this technique are that it can be employed to achieve particle drying by which the core materials in a liquid form can be converted into a dry powder form without additional drying and the products offer direct tableting characteristics because of their agglomerated structure and thus offer significant advantages to the candy and pharmaceutical industries [83].
2.7.11 Spray Drying

Spray drying has been widely utilized for commercial production of powders of fruits and vegetables. Spray-dried powders have good reconstitution characteristics, low water activity and are suitable for transport and storage. Furthermore, it has been successfully applied for heat sensitive components such as carotenoid stability in plant foods such as carrots, tomato pulp, sweet potato and sea buckthorn [121 – 124].

Spray drying is one of the oldest processes to encapsulate aroma. It is so common in foods to use aroma in a spray-dried, fine powder form that one often tends to forget it is a form of encapsulation. About 80–90% of encapsulates are spray-dried, 5–10% are prepared by spray-chilling, 2–3% are prepared by melt extrusion and 2% are prepared by melt injection [125]. Other type of encapsulates contribute <1% to all encapsulates.

Spray drying of aroma is achieved by dispersion of the aroma in an aqueous solution of carrier material, followed by atomization and spraying of the emulsion into a hot chamber. The co-current, hot air stream has a predetermined temperature within the 160–220°C range, which heats the droplets almost instantaneously to 100°C [126]. During this process a film is formed at the droplet surface and the concentration of ingredients in the drying droplet keeps increasing, thereby retarding the larger aroma molecules while the smaller water molecules continue to diffuse to the exterior at a considerable rate [127, 128]. The final powder consists of dry particles containing a dispersion of fine aroma droplets and having a particle size of 10–150 µm.

Spray drying has been used for many years to encapsulate various kinds of food ingredients [129]. A liquid solution or suspension is passed through a small nozzle, which leads to the formation of a mist of fine drops. The outlet of the nozzle is located in controlled high temperature environment so that the volatile liquid phase (usually water) within the drops quickly evaporates. Spray driers typically operate at temperatures between 150 and 300°C depending on the nature of the material being
prepared. The actual temperature experienced by the material within the drops is considerably less than this because of the latent heat associated with liquid evaporation. In addition, the high surface-to-volume ratio of the drops allows for rapid drying, which also minimizes thermal damage. Spray drying is capable of continuous operation and produces a dry powdered product. The diameters of the particles within the spray-dried powder are usually in the 10-100 µm range, so that each particle usually contains many different lipid droplets or biopolymer molecules [126]. Spray drying can be used to convert a suspension of biopolymer particles into a powder that can be reconstituted prior to use.

Spray drying is the most widely used industrial process for removal of solvent involving particle formation and drying. One problem in spray drying is the stickiness. To minimize the stickiness problem, both process-based and material science-based approaches are in place. Process-based approaches include the mechanical scraping of the chamber wall, the introduction of cold air at the bottom, and the use of low temperature/low humidity air. An example of the material science-based approach involves the addition of drying agents to reduce the stickiness of the powders [130]. Several researchers have added drying aids which are high molecular weight carbohydrates such as maltodextrin to reduce the stickiness of the material and reduce wall deposition problems.

Various spray drying conditions such as product feed rate, air flow, feed temperature, inlet air temperature, and outlet air temperature need to be optimized in order to produce discrete well-formed microspheres [131].

Retention of volatile core material during encapsulation by spray drying is achieved by chemical and physical properties of the wall and core materials [132], solid content of the dryer, processing temperature and also by the nature and the performance of the encapsulating support, i.e. emulsion-stabilizing capabilities, film-forming ability and low viscosity at a high concentration [133]. The functionality profile of wall
materials that are optimal for spray drying includes a high solubility in water, a low viscosity at high concentration, effective emulsification and film-forming characteristics and efficient drying properties [23]. When core materials of limited water solubility are encapsulated by spray drying, the resulting capsules are of a matrix-type structure. As such, the core is organized into small droplets coated with wall materials that are embedded in the wall matrix. Microstructures of spray-dried capsules have been shown to be affected by wall composition and properties, core-to-wall ratio, atomization and drying parameters, uneven shrinkage at early stages of drying, the effect of a surface tension-driven viscous flow and storage conditions [134, 135].

Borrmann et al., [136] found that the microencapsulated passion fruit juice powder with $n$-octenylsuccinate-derivatised starch as carrier retained 77.1 and 71.5%, of vitamin C after 77 days of storage when stored at 7 and 25°C which proved spray-drying itself as an inexpensive alternative to freeze-drying capable of retaining vitamin C during a long time of storage, and it was easy to be diluted in order to reconstitute the passion fruit juice for human consumption.

2.8 SELECTION OF THE WALL MATERIAL

The most important criteria for selection of an encapsulation material are functionality that encapsulate should provide to the final product, potential restrictions for the coating material, concentration of encapsulates, type of release, stability requirements and cost constraints. Materials used for design of protective shell of encapsulates must be food-grade, biodegradable and able to form a barrier between the internal phase and its surroundings. The majority of materials used for encapsulation in the food sector are biomolecules. Selected wall materials have to provide maximal protection of the active material against environmental conditions, to hold actives within capsules structure during processing or storage under various conditions, not to react with the encapsulated material, to have good rheological characteristics at high concentration if it is needed and to have easy work ability during the encapsulation [86].
Among all materials, the most widely used for encapsulation in food applications are polysaccharides. Starch and their derivatives – amylose, amylopectin, dextrins, maltodextrins, polydextrose, syrups and cellulose and their derivatives are commonly used. Plant exudates and extracts – gum arabic, gum tragacanth, gum karaya, mesquite gum, galactomannans, pectins and soluble soybean polysaccharides are employed, too. Subsequently, marine extracts such as carrageenans and alginate are also present in foods. Microbial and animal polysaccharides like dextran, chitosan, xanthan and gellan are also exploited. Apart from natural and modified polysaccharides, proteins and lipids are also appropriate for encapsulation [86]. Examples of the most common milk and whey proteins are caseins, gelatine and gluten. Among lipid materials suitable for food applications there are fatty acids and fatty alcohols, waxes (beeswax, carnauba wax, candellia wax), glycerides and phospholipids. In addition to the above, other materials are employed such as poly vinyl pyrrolidone (PVP), paraffin, shellac and inorganic materials [81].

The carrier material of spray-dried aroma should ideally have a high degree of solubility, limited viscosity at 35–45% (w/w) in water, good emulsifying properties, good film forming and drying properties, a non-hydroscopic character, a bland taste, should be non-reactive, should protect the aroma from oxidation and should be available at low cost [125, 128].

Gums are used in microencapsulation for both film forming and emulsion stabilization properties. Among all gums, acacia gum, generally called gum arabic, stands out due to its excellent emulsification properties and thus is widely used. Gum arabic is a polymer consisting of D-glucuronic acid, L-rhamnose, D-galactose, and L-arabinose, with approximately 2% protein [137]. The emulsification properties of the gum arabic are attributed to the presence of this protein fraction [138].

Gum arabic is a hydrocolloid produced by natural extrudate from the trunk and branches of leguminous plants of the family Acacia. It is one of the oldest, traditional
and effective encapsulation agents used in spray drying. It is odourless, colourless, tasteless and does not affect the odour, colour, and taste of the system to which it is added [81]. It possesses excellent solubility in water and surface active properties, and produces low-viscosity solutions at high solids concentrations [139].

Although it presents many desirable characteristics to be a good encapsulating agent (high solubility, low viscosity and good emulsifying properties), the oscillation in supply, as well as the increasing prices, is leading researches to look for alternative wall materials that could replace it or be used in combination with it [140].

Mixture of gum arabic and maltodextrin was reported effective in microencapsulation of cardamom oil using spray drier [141], capsicum oleoresin [142], soy oil [143]. Microencapsulation of red pepper oleoresin using gum arabic and modified starch has also been tried by Jung and Sung [144].

Maltodextrins are non sweet, creamy white and nutritive saccharide polymer \((\text{C}_6\text{H}_{10}\text{O}_5)_n\text{H}_2\text{O}\) which are obtained by acid hydrolysis of several starches (corn, potato or others). They differ in average molecular size (DEs: 4, 10, 15, 20, 25, 30, and 42) [145] and are classified on the basis of dextrose equivalence (DE), which is a measure of the degree of starch polymer hydrolysis. Maltodextrin products will have various chain lengths of polymer of the anhydroglucose unit. The average molecular weight decreases when DE increases. For example, the average molecular weight of maltodextrin DE5, DE10, DE15 are 3600, 1800 and 1200, respectively. Maltodextrin from different botanical source exhibited different characteristics. Rice maltodextrin consisted of lower molecular-weight saccharides while potato maltodextrin had higher molecular-weight saccharides. It is generally recognized as safe and can be used directly as a food ingredient [146].

In general, maltodextrins have high solubility in water, low viscosity, bland flavour, colourless solutions [147] and are extensively used in the food industry. It is a filler matrix [135], which is cheap, highly soluble in water, able to form stable emulsion.
and easily digestible [81]. The reason of choosing maltodextrin is that they protect encapsulated material from oxidation [149] by forming amorphous glassy matrices during the encapsulation process [150], thereby it increases encapsulation stability [151]. It is also mainly used to reduce stickiness and agglomeration problems during storage, thereby improving product stability [152].

A maltodextrins characterization study published by Raja et al., [153] showed that maltodextrins with dextrose equivalence between 10 and 20 are fit for using as wall material. Those maltodextrin samples show the highest retention of flavor because they could be dispersed in water up to 35.5% of the solution without haze formation.

Maltodextrin DE 18.5 was considered as the most suitable partial replacer for gum arabic because it presents a good solubility and a rapid reconstitution of the emulsion in water [143].

Currently, maltodextrin is one of the common drying aids for spray drying owing to its beneficial role as a carrier or an encapsulating agent in increasing the stability of carotenoids, reasonably cheap and commercially available. The addition of maltodextrin before spray drying has been reported to be effective in preserving carotenoids such as β-carotene [154]; paprika oleoresin [155], carrot carotenes [121]; blackcurrant, apricot and raspberry juices [153]; guava juice [156] and pineapple juice [157]. Cai and Corke [146] observed that the spray-dried powders with maltodextrin had better solubility and dispersibility.

2.9 EMULSION PREPARATION

One of the key steps in the encapsulation of oils and flavours by spray drying is the emulsion preparation, which plays an important role in the surface oil content present in the final encapsulated powder. The significant parameters to be considered in the emulsion formation are: total solid concentration, oil content, viscosity, stability, droplet size and emulsification method. If the emulsion is sufficiently stable and
presents optimal conditions of viscosity and droplet size, then the encapsulation efficiency can be maximized by the right choice of the spray drying parameters, including inlet and outlet air temperatures, emulsion temperature, atomization conditions, drying air flow rate and humidity [127]. Thus, it is also important to optimize the drying process, in order to obtain the minimal surface oil in the powdered particles.

In a reported study, the wall material (gum arabic) was completely dissolved in distilled water, under magnetic stirring. Coarse emulsions were prepared by blending the coffee oil and the wall solution, using a rotor-stator homogenizer (Ultra-turrax IKA T18 Basic, Wilmington, NC, USA) operating at 14,000 rpm for 5 min. Then the emulsion is subjected to spray drying process to produce microencapsulated coffee oil powder [158].

Thirty percent w/v solution of the different blends of gum arabic, maltodextrin and the commercial modified starch i.e. HiCap® 100 were dispersed in distilled water and final volume made to 100ml. It was rehydrated for about 12 h at refrigerated temperature (10–12°C). Three grams (10% based on the carrier used) of oleoresin was added to the mixture. The mixture was emulsified in a shear homogenizer (Indofrench Industries Engineers, Mumbai, Model type- SPM-9) for 5min at 3000 rpm until complete dispersion of the oleoresin. Two drops of Tween 80 was added for proper emulsification. The resulting slurry was spray dried in JISL LSD-48 mini spray dryer [30].

### 2.10 SIGNIFICANT PARAMETERS INFLUENCING EMULSION CHARACTERISTICS

The emulsion characteristics such as emulsion stability, viscosity, particle size in emulsion, feed concentration, ratio of core to wall material are discussed below.
2.10.1 Emulsion Stability

Barbosa et al. [159] stated that the more stable emulsion produced the higher encapsulation efficiency i.e. the lower the amount of non-encapsulated material on particles surface.

Many studies have shown that the reduction of emulsion droplets size, which generally represents an increased stability, results in greater retention of active material. In general, small emulsion droplets will be enclosed and embedded more efficiently within the wall matrix of the microcapsules and the resulted emulsion will be more stable during the spray drying encapsulation process, which is one of the critical parameters to have the optimum efficiency [160,161].

Carneiro et al., [162] analysed the emulsion stability on flaxseed oil using different combinations of wall materials. This study revealed that most of the emulsions were kinetically stable, with exception of those prepared with whey protein concentrate and maltodextrin, which showed the formation of a small separation layer and a foam phase, 24 h after its homogenization. This was unexpected, since whey proteins are well known by their good emulsifying capacity. According to Dickinson and Matsumura [163], this result may have been caused by the unfolding of the protein molecules at the droplets surface, which would enhance protein–protein interaction leading to flocculation during emulsification and consequently reducing the emulsion stability. The unfolding of protein molecules of the oil–water interface may lead to changes in secondary and tertiary structure, and consequently exposure of their residues which would be linked (–S–S– linkages or disulphide linkages) within the native globular structure leading to the formation of intermolecular interaction at the oil–water interface and flocculating. Another hypothesis that can be considered to explain this unusual behaviour is that the stability of protein-stabilized emulsions is a function of pH and other parameters. So, depending on the emulsion pH, the emulsifying capacity of whey
protein concentrate may have been lower than usual [164], affecting the emulsion stability.

2.10.2 Emulsion Viscosity

In a feed emulsion viscosity exerts an effect on flavour retention during spray drying by the circulation currents within the drying droplet and the time to form discrete droplets [165]. If the viscosity is low, internal mixing occurs during drying which delays the formation of a semi permeable surface. This delay permits greater flavour loss during early drying, thus an increase in feed viscosity should favour volatile retention. However, increasing the viscosity too much will slow the formation of discrete particles during atomization which promotes volatile losses [166].

Hayashi and Kudo, [167] results demonstrated that the viscosity of milk increased exponentially with an increase in feed concentration. The higher the feed solid content and the lower the oil concentration, the higher was the emulsion viscosity. Emulsions with higher viscosity require shorter time to form a crust, thus reducing the circulation movements inside the droplets and resulting in higher oil retention [127].

Hogan et al., [168] informed that the increase in apparent viscosity was most likely attributed to an increase in the core material concentration and emulsion total solids concentration.

According to Masters [169], the atomized droplet size varies directly with emulsion viscosity at a constant atomization speed. The higher the emulsion viscosity, the larger are the droplets formed during atomization, and therefore, the larger are the powdered particles obtained. Carneiro et al., [162] indicated that droplets size was not affected only by the emulsion viscosity, but also by the intrinsic emulsifying properties of each type of material.
Kshirsagar et al. [24] observed that the viscosity increased slightly with an increase in oleoresin loading, probably due to the greater tendency of the oleoresin to form agglomerates at higher levels.

2.10.3 Particle Size in Emulsion

Many research studies revealed that there was a direct relationship between emulsion droplet size and encapsulation efficiency. The lower was the particle size, the higher was the efficiency [160, 170]. In general, small emulsion droplets will be enclosed and embedded more efficiently within the wall matrix of the microcapsules and the resulted emulsion will be more stable during the spray drying encapsulation process, which is one of the critical parameters to have optimum efficiency [171]. In addition, according to Soottitantawat et al. [172], the larger emulsion droplets would be sheared into smaller droplets because of the large velocity gradient and the turbulence in the thin liquid film on the surface of the atomizer. However, a lower emulsion size does not necessarily correspond to a longer shelf-life, since the greater surface area of the oil droplets embedded in the capsule wall provides greater possibility for oxidation once oxygen has penetrated into the particle [22].

2.10.4 Ratio of Core to Wall Material

Concerning the ratio of core to wall material, many authors refer that using the highest possible core concentration that provides high core retention in microcapsules is advantageous, because less wall material is needed, leading to increased yield and output, with positive economic impact [127]. However, in most of the published studies, a typical core to wall material ratio of 1/4 is adopted and reported as being optimal for various wall materials, like gum arabic and modified starches [83]. Higher oil loads resulted in higher surface oil content of the powder and lower encapsulation efficiency [158,173]. This trend can be attributed to greater proportions of core materials close to the drying surface, thereby shortening the diffusion path length to the air/particle interface [127]. The influence of core to wall material ratio on efficiency can also be
related to the emulsion viscosity, since lower oil load results in higher emulsion viscosity [158], which makes difficult the oil diffusion to the particle surface.

2.10.5 Feed Concentration

Cai and Corke [145] found that increasing feed solid content (10.5% to 39.2%) could raise powder productivity significantly at the same air flow rate, compressor air pressure and inlet-air temperature (180°C), which could reduce production cost. Also increase in the feed solid content, the bulk density was also increased.

The highest possible in feed solids content should be used, since high solids content reduces the required time to form a semi-permeable membrane at the surface of the drying particle [174]. A fast formation of a solid surface could be associated with low levels of surface oil content as there is less opportunity for the core material droplets to come onto the particles surface [161] and lead to an increase in encapsulation efficiency [173].

In addition, the higher the total solids, the higher the emulsion viscosity, thus preventing the circulation movement inside the droplets, thereby resulting in a rapid skin formation [127]. However, other researchers suggest that there is an optimum in feed solids content [175] which can be attributed to two reasons: first at some solids content, adding more wall material exceeds its solubility and these undissolved materials cannot provide any effective encapsulating effect. Second is related to the larger exposure during atomization, the slow formation of discrete droplets during atomization and the difficulties in droplet formation [176]. Too high an infeed viscosity delays particle formation during atomization which tends to favour volatile losses during drying. It is apparent that each carrier material has its own optimum in feed solids for flavour retention which is primarily based on solubility and viscosity in solution [177]. The emulsions produced with higher solid content showed lower droplet mean diameters, which resulted in lower encapsulation efficiency. The lower the
encapsulation efficiency, the higher the amount of surface oil, which was more exposed to the drying air and thus, more susceptible to evaporation and loss of volatiles.

2.11 SIGNIFICANT PARAMETERS INFLUENCING SPRAY DRYING PROCESS

2.11.1 Air Flow Rate

The effect of drying air flow rate on powder bulk density depends on its effect on moisture content, as a product of higher moisture would tend to have a higher bulking weight caused by the presence of water, which is considerably denser than the dry solid [178]. As a result, air flow rate increases led to an increase in powder moisture content and an increase in bulk density [173].

As far as the drying air flow rate effect is concerned, the movement of air predetermines the rate and degree of droplet evaporation by influencing (a) the passage of spray through the drying zone, (b) the concentration of product in the region of the dryer walls, and (c) the extent to which semi-dried droplets re-enter the hot areas around the air disperser. A lower drying air flow rate, causes an increase in product sojourn time in the drying chamber and enforces circulation effects [179]. Increased residence times lead to a greater degree of moisture removal. As a result, an increase in drying air flow rate, decreasing the residence time of the product in the drying chamber, led to higher moisture contents [173].

2.11.2 Feed Temperature

In fact, feed temperature modifies the viscosity of the emulsion, its fluidity and thus, its capacity to be homogenously sprayed. When the feed temperature is increased, viscosity and droplets size should be decreased but high temperatures can cause volatilization or degradation of some heat-sensitive ingredients [180].
Shu et al., [181] conducted experiments on encapsulation of lycopene by spray-drying and it was observed that increasing feed temperature from 35 to 65°C while keeping other parameters unchanged, both encapsulation yield (EY) and encapsulation efficiency (EE) increased at the beginning and reached a high value of 91.2%, 81.3%, respectively, as the feed temperature reached 55°C. When feed temperature increased to 65°C, EY and EE were decreased both. Increasing feed temperature led to low viscosity of emulsions, which may result in a good atomization effect when spray-drying, thereby a higher EY and EE; but when feed temperature was too high (reached 65°C), a lot of particles accumulated on the chamber wall were found and a burnt smell was dispersed and low EY and EE were obtained. Therefore, the optimal feed temperature of 55°C was determined.

The feed temperature adjustment is crucial to modify the viscosity of the polymer solution and in turn, its capacity to be sprayed homogeneously [93].

2.11.3 Feed Rate

The rate of feed delivered to the atomizer is adjusted to ensure that each sprayed droplet reaches the desired drying level before it comes in contact with the surface of the drying chamber. Moreover, appropriate adjustment of the air inlet temperature and flow rate is important [180]. Chegini and Ghobadian [178] concluded that increase in feed flow rate, decreases the average time of wettability and insoluble solids of powder.

Laohasongkram et al., [182] observed that increasing feed rate resulted in the increases in particle size, moisture content, bulk density, percentage of microencapsulation efficiency but a decrease in surface oil in encapsulation of macadamia oil using spray drying technology.
2.11.4 Inlet Air Temperature

The air inlet temperature is usually determined by two factors: the temperature which can safely be used without damaging the product or creating operating hazards and the comparative cost of heat sources [183].

Reineccius and Coulter [184] reported that inlet air temperatures of 160 to 210°C give optimum flavour retention during spray drying encapsulation of various products and also suggested that inlet air temperatures above 210°C have been found to decrease flavour retention.

In fact, air inlet temperature is directly proportional to the microcapsule drying rate and the final water content. When the air inlet temperature is low, the low evaporation rate causes the formation of microcapsules with high density membranes, high water content, poor fluidity, and easiness of agglomeration. However, a high air inlet temperature causes an excessive evaporation and results in cracks in the membrane inducing subsequent premature release and a degradation of encapsulated ingredient and also a loss of volatiles [185]. A higher evaporation rate can be produced when the feed spray passes through a lower humidity and/or higher temperature drying chamber and produce finer particles during spray drying [186].

Walton and Mumford [187] informed that the greater the inlet drying temperature, the greater the thermal efficiency of the process. However, this must be balanced against the thermal stability of the product.

According to Walton [188], an increase in the drying air temperature generally produces a decrease in bulk and particle density, and there is a greater tendency for the particles to be hollow. The former can be caused by particle inflation-ballooning or puffing, and is particularly common in skin-forming materials such as gum arabic, rice starch, wheat starch or dextrin [81,86].
Shiga et al., [189] reported higher retention of citral / linalyl acetate and shiitake flavor, respectively, at higher drying temperatures. On the contrary, Aburto et al., [190] and Finney et al., [191] showed that the retention of encapsulated material was independent of the air temperature.

An increase in inlet drying temperature, resulted in a greater loss of lycopene content in tomato powders [122] and watermelon powder [192] due to thermal degradation and oxidation.

For spray drying in general, increasing drying temperature resulted in greater loss of water of resultant powder, due to the higher rate of heat transfer into particles, causing faster water removal. Also it was discussed that in increasing the drying temperature from 120°C to 200°C significant loss of total antioxidant activity (TAA) was observed, from 0.14 to 0.08 mmole Trolox equivalents/g of powder. However, there was no statistical difference in TAA of samples spray-dried at temperatures of 140°C and 160°C [193].

The use of high temperatures has also been reported to severely impact the viability of encapsulated cells due to dehydration of the cells as well as the inactivation of essential enzymes that maintain cellular balance [194].

Goula and Adamopoulos [173] observed that air inlet temperature is directly related to the microcapsule drying rate and the final water content. A high enough inlet air temperature leads to a rapid formation of the semi-permeable membrane on the droplet surface, giving optimum core material retention. A higher temperature could cause heat damage to the dry product or “ballooning” and excessive bubble growth with surface imperfections, which increase losses during spray drying.

Decrease of loose bulk density with a temperature increase from 120 to 160°C was observed by Chegini and Ghobadian [178] in the case of orange juice microencapsulation, and by Fazaeli et al., [195] for black mulberry juice which is
mainly due to the higher evaporation rates which can produce more porous powders with lower shrinkage of the droplets during drying.

Chegini and Ghobadian [196] observed that the β-carotene content in the orange juice powder decreased with increasing inlet air temperature with maltodextrin as the carrier. The powder was characterized by a high content of pigments, although it decreased compared to the starting juice. The inlet air temperature of 140°C could be recommended as a good condition because of high content of betanin pigments and also because of good physical properties of microcapsules (particle size, porosity, apparent particles density, loose density and dry matter content).

2.11.5 Outlet Air Temperature

In a spray drying system, the moisture content is controlled by the temperature of the exhaust air leaving the drying chamber. The humidity of the air may also be a factor. High ambient air humidity may require an increase in outlet air temperature in order to maintain the desired powder moisture content [197]. The residual moisture in the powder influences many other powder properties such as bulk density, solubility and flow behavior [198,199].

2.12 EFFECT OF PROCESS PARAMETERS ON QUALITY ANALYSES

2.12.1 Bulk Density

Bulk density is important in packaging and shipping considerations. Absolute density, particle geometry, shape and size influence bulk density. Spherical particles pack the best and thus, have the highest bulk densities, all other factors being equal [176].

The apparent density increased with the increase of the feed flow rate and with the decrease of the atomization speed [178, 200], since higher feed flow rate lead to a
minor degree of moisture removal, and this moisture content, due to higher density compared to the dry solid, contributed to the increase of this property. At lower atomization speed the particles become larger and denser [201].

Bulk densities of tuna flavour powder produced from tuna concentrate with maltodextrin of 20, 22, 24 and 26% total soluble solids (TSS) were 0.39±0.01, 0.41±0.01, 0.41±0.01, and 0.44±0.02 g/ml, respectively [202]. Bulk densities of the powder from tuna concentrate with 20, 22 and 24 % TSS were significantly lower (p<0.05) than from 26% TSS. Hence, it was observed that the bulk density of the spray dried tuna flavour powder was increased with increasing maltodextrin levels [202]. The more moisture content of the sample causes high bulk density which implied that the particles tend to stick together [203].

The porous structures are favored by high drying rate promoted by the use of high temperatures due to the expansion of evaporation of water vapour leaving the empty spaces occupied by the air [204]. According to Walton, [188] increasing the drying air temperature generally produces a decrease in bulk and particle density, and there is a greater tendency for the particles to be hollow. The former can be caused by particle inflation-ballooning or puffing, and is particularly common in skin-forming materials.

Kha et al., [193] observed that the bulk density of Gac (Momordica cochinichinensis) fruit aril powders was significantly affected by the drying temperature (p < 0.01), with decreasing density observed with increased drying temperature. At very high temperatures, very high drying processes are achieved implying a lower shrinkage of the droplets, and so a lower density of the powder [205].

Carneiro et al., [162] obtained higher bulk density value in the formulation of maltodextrin and gum arabic than combinations of maltodextrin with modified starch -capsul, Hi-cap and whey protein concentrate. The advantage of obtaining powders with higher density is that they can be stored in large amounts into smaller containers when
compared to products with lower densities. Moreover, higher bulk density may indicate lower amount of air occluded in the spaces between particles, which can help to prevent lipid oxidation.

### 2.12.2 Moisture Content

Moisture content is the important quality parameter since it affects the powder packing, particle agglomeration, flowability and bulk density [187].

Dian et al., [206] found moisture content values varying from 2.2 to 3.0% for microencapsulated palm oil and observed that this property was not affected by the type of wall material.

Hogan et al., [168] observed moisture content values from 1 to 3% in soybean oil microencapsulated by spray-drying and these values were not affected by the type of wall material.

Goula et al., [198] reported that increasing residual moisture content of tomato powder decreased its bulk density. However, that trend was due to the thermoplastic nature of the product.

Goula and Adamopoulos [199] reported that an increase in air inlet temperature leads to a decrease in moisture content. The greater the temperature difference between the drying medium and the particles, the greater will be the rate of heat transfer into the particles, which provides the driving force for moisture removal. When the drying medium is air, temperature plays a second important role. As water is driven from the particles in the form of water vapor, it must be carried away, or the moisture will create a saturated atmosphere at the particle surface. This will slow down the rate of subsequent water removal. The hotter the air, the more moisture it will hold before becoming saturated. Thus, high temperature air in the vicinity of the drying particles
will take up the moisture being driven from the food to a greater extent than with cooler air.

Fernandes et al., [207], studying the microencapsulation of *Lippia sidoides* essential oil, observed that moisture content was reduced from 5% to 4% as a consequence of an increase in the total solids from 30% to 60%.

As maltodextrin concentration increased from 10% to 20%, the moisture content of samples significantly reduced from 4.87% to 4.06% and increasing drying temperatures from 120°C to 200°C, resulting in a significant drop in moisture content from 5.29% to 3.88% in Gac fruit aril powder [193]. The higher moisture content was obtained by powder spray drying at lower inlet air temperatures. Increasing moisture content caused a higher loss of lycopene. However, when moisture content increased, a greater degree of aggregation occurred because of the natural stickiness of the product. As the sample is embedded, it leads to lower oxygen exposure resulting in lower lycopene loss [198].

Kanpairo et al., [202] utilized the waste from canned tuna processing (tuna precooking juice) for producing dried tuna flavour powder. Tuna precooking juice was centrifuged and concentrated to 15% total soluble solid (TSS). Maltodextrin (DE 9) was added to increase the TSS of tuna precooking concentrate to 20, 22, 24 and 26%, then dried by spray dryer at 180°C inlet air temperature. The moisture content of the spray dried tuna flavour powder ranged from 4.63±0.2 to 7.46±0.3%. Moisture, protein, salt and lipid contents were observed to decrease with increase in the TSS of maltodextrin and decrease in ash content and percentage yield of the powder.

### 2.12.3 Solubility

Solubility is one of the most important physicochemical and functional properties of concentrates. Solubility of the tomato powders varied from 121 to 245 s. The effect of drying air flow rate on powder solubility depends on its effect on powder
moisture content, as low moisture content seems to be associated with fast dissolution. Air flow rate increases lead to an increase in powder moisture content and a decrease in powder solubility [208]. The time required for the powder to dissolve was found to increase with an increase in compressed air flow rate, since particle size affects solubility rate. Large particles may sink, whereas small ones are more dusty and generally float on water resulting in uneven wetting and reconstitution [209].

Solubility of tuna flavour powder ranged from 60.87 to 70.12%. The solubility increase with increase in maltodextrin level up to 24% TSS [202]. There seems to exist a tendency for water solubility increased when increasing replacement of maltodextrin [210]. The high solubility of powder indicated potential applications in formulated food systems by providing an attractive appearance and a smooth mouth feel to the product [211].

While assessing water-solubility, Wang et al., [212] reported that the free lutein could not be dissolved with water as solvent at room temperature while spray dried encapsulated lutein could be dissolved immediately after 120 s. There was no deposit in the solution, whose colour and luster of solution were vivid and transparent. Porous starch improved the absorbability and adhesive property of the material. It made all material well dispersed, so the solubility of encapsulated lutein might be improved.

### 2.12.4 Water Activity

Karel and Langer [213] indicated that the water activity \( a_w \) rather than water content determines the lower limit of available water for microbial growth. Most bacteria don’t grow below \( a_w \) of 0.91; most moulds ceased to grow below \( a_w \) of 0.80 and the lowest limit for xerophilic fungi growth was at 0.65. It was also reported that the \( a_w \) modified the sensitivity of microorganisms to heat, light and chemicals. They are more sensitive at higher \( a_w \) and minimum sensitivity occurs at intermediate level highlighting pronounced effect of synergic system.
Quek et al., [192] stated that the $a_w$ of spray-dried water melon powders was not significantly changed by inlet temperatures of between 145°C and 175°C. Further, higher concentration of maltodextrin resulted in a decrease in the $a_w$ of the powders.

Kanpairo et al., [202] reported the $a_w$ of tuna flavour powder decreased from 0.48±0.02 to 0.33±0.01 with increasing levels of maltodextrin from 20 to 26 % TSS.

2.12.5 Colour

The increased drying temperatures resulted in low retention in the redness colour of carrot products [214] and of tomato products [215]. The possible explanation for this phenomenon is that carrying out the spray-drying process with a high ratio of surface area and volume of feed mixture caused rapid pigment oxidation [154]. Therefore, the spray drying conditions at high temperature resulted in a high loss of red colour due to the thermal degradation of carotenoid.

Goula and Adamopoulos [22] indicated that a higher loss of lycopene content in tomato powder was observed by increasing the air inlet temperature. In terms of the maltodextrin concentration, moreover, the lesser redness of Gac fruit powders was due to the higher concentration of maltodextrin used in the spray-drying process [193]. Grabowski et al., [123] found that increasing maltodextrin resulted in an increase in hue angle in sweet potato powders, indicating a loss of redness.

Sousa et al., [216] found that the highest value of lightness of spray-dried tomato powders was observed at the highest inlet drying temperature, indicating less darkness due to the pigment oxidation. In contrast, the lightness of water melon powders reduced when inlet drying temperature increased due to the high content of sugar causing browning of powders [192]. An increase in the lightness of products was significantly obtained by increasing maltodextrin concentration from 10% to 20% [193].
2.12.6 Encapsulation Efficiency

The encapsulation efficiency (EE) reflects the presence of free oil on the surface of the particles within the powder and the degree to which the wall matrix can prevent extraction of internal oil through a leaching process [168].

Beristain et al., [217] analyzed that the effect of increasing initial concentration of the emulsion from 200 to 250 g of cardamom essential oil/kg of gum which probably causes an increase in the concentration of oil in the wall and outer surface of the capsule during crust formation and more oil is lost [132]. Such losses are enhanced by internal circulation streams inside the drying liquid during droplet formation in spray drying and the expansion of droplets followed by crater formation in the dry skin, leading to evaporation from the interior of the particle [218].

Tan et al., [219] found that an increase in oil loading resulted in lower microencapsulation efficiency, with less oil being encapsulated. This could also be due to the lower amount of wall material used to encapsulate the oil as the oil load was increased [220].

Zhu et al., [221] conducted experiments for the encapsulation of lycopene pigment by spray-drying method using gum arabic and modified starch as wall materials. It was found that increasing homogenization pressure from 0 to 60 MPa, while keeping other parameters unchanged, increased the encapsulation efficiency dramatically at the beginning and then gradually tended to a plateau until the pressure reached 60 MPa.

Hogan et al., [222] found that by increasing soy oil-sodium caseinate ratio from 0.25 to 3.0, microencapsulation efficiency dramatically decreased from 89.2 to 18.8% during spray drying. This general trend is attributed to greater proportions of core materials, particularly volatiles, close to the drying surface, thereby shortening the diffusion path length to the air /particle interface.
The surface oil can be easily oxidized to form off-flavour compounds. The amount of surface oil in the spray-dried powder is quite important for stable storage [172].

According to Barbosa et al., [159], the more stable the emulsion, the higher the encapsulation efficiency, i.e., the lower the amount of nonencapsulated material on particles surface.

Charve and Reineccius [140] obtained a result while studying the volatile retention in microcapsules prepared by spray drying, where the microencapsulated particles produced with modified starch showed higher oil retention when compared to the particles encapsulated with gum arabic and with whey protein. Many studies have shown that the reduction of emulsion droplets size, which generally represents an increased stability, results in greater retention of active material [32, 160, 161].

The encapsulation efficiency of the Gau fruit aril powder was also significantly influenced by maltodextrin concentration and by drying temperature (p < 0.001). Increasing maltodextrin concentration resulted in higher encapsulation efficiency however, no difference in encapsulation efficiency between the concentration of 20% and 30% was observed. Moreover, in general, encapsulation efficiency of the samples reduced from 76.6% to 48.0% as the drying temperature increased from 120°C to 200°C, respectively [193].

Frascareli et al., [158] found that encapsulation efficiency and oil retention were negatively influenced by oil concentration and inlet air temperature in microencapsulation of coffee oil by spray drying.

Goula and Adamopoulos [173] studied the effect of drying air flow rate on encapsulation efficiency. It was observed that increasing drying air flow rate, thus increasing outlet air temperature, encapsulation efficiency increased. This observation can be attributed to a lower relative humidity at higher outlet air temperatures, which
results in more rapid drying, whereas Anker and Reineccius [223] reported that increasing outlet air temperature results in higher surface oil contents of particles, probably due to the “ballooning” effect, where particles may develop fissures and even split. However, that effect was reported mainly for volatile core materials.

Carneiro et al., [162] inferred that the encapsulation efficiency of samples was significantly influenced by the type of wall material used, since emulsions prepared with Hi-Cap resulted in particles with considerably lower surface oil than those prepared with maltodextrin (MD) and whey protein concentrate (WPC). The encapsulation efficiency values varied from 62.3% to 95.7%, being the lowest value obtained for MD: WPC. Analyzing the results, these two combinations (MD: WPC and MD:Hi-Cap) had similar rheology and droplet mean diameter characteristics. However, the emulsion containing MD and WPC showed the poorest stability, which might have influenced the encapsulation efficiency.

2.12.7 Allicin

Koch et al., [224] described a simple, fast and reliable procedure for determining allicin in garlic bulbs and in its preparations by static headspace gas chromatography. After enzymatic release, the allicin was quantitatively reduced by Cleland's reagent (1,4-dimercapto-2,3-butanediol) without prior isolation. The allylmercaptan formed was sampled from the headspace above the aqueous dispersion and injected into the gas chromatography. Quantification was achieved either by addition of a suitable internal standard, e.g. 1-pentylmercaptan, or by using a homogenized garlic powder as external standard.

Lawson et al. [225] employed high performance liquid chromatography (HPLC) method to quantify allicin, and other thiosulfinates in garlic clove homogenates. The method reported was tedious and requires expensive instruments and hence Wanyika et al., [226] suggested a fast and cheap method for assaying allicin, the active constituent of garlic extract, based on UV spectrophotometry. In this method, garlic cloves were
extracted using water. The allicin content of the garlic extracts was analysed after passing the extract through Solid Phase Extraction (SPE) cartridge and eluted with solvents of various polarities. The most polar solvent used was water which eluted the allicin in 4 ml, while methanol and ethanol did not. The absorbance of UV radiation at 240 nm and 254 nm wavelengths by the garlic fraction eluted with water gave a ratio of A240 nm/ A254 nm, 1.4 – 1.5, which is typical for allicin

HPLC, gas chromatography (GC), HPLC–mass spectroscopy (HPLC-MS) and GC–MS can be used for determination of various components of garlic and aged garlic extract, including allicin [227].

2.13 QUALITY ANALYSES OF THE ENCAPSULATED POWDER

2.13.1 Ash Content

In the production of powders, the ash content may be affected. The ash content of the tuna flavour powder was increased from 3.44 ± 0.3 to 4.25 ±0.3% when the total soluble solids was raised from 20 to 26% using maltodextrin. Hence it is inferred that addition of maltodextrin to tuna precooking concentrate not only increased the total soluble solids but also ash content of the powder [202].

2.13.2 pH

The values of pH of the Gac fruit aril powders [193] and Roselle extract powders [228] were not significantly affected by inlet air drying temperature and also by maltodextrin concentration (p > 0.05) [193].

2.13.3 Total Antioxidant Activity

Antioxidants are considered important nutraceuticals on account of many health benefits. Tonon et al., [229] found that antioxidant activity of spray-dried acai (Euterpe oleracea Mart.) juice produced with maltodextrins and gum arabic did not significantly
differ between each other, while the powder produced with tapioca starch showed the 
lowest DPPH scavenging capacity after the spray drying process. Also it was reported 
that antioxidant activity decreased with increasing water activity, which is related to the 
higher anthocyanin degradation caused by moisture. Temperature, however, exhibited 
an unexpected effect, a higher antioxidant activity was observed in the samples stored at 
35°C, when compared to 25°C which might be due to the presence of compounds in 
açaí, other than polyphenolics, that contribute to its antioxidant capacity, and the 
occurrence of Maillard reaction.

Krishnaiah et al., [230] suggested that loss of bioactive components can be 
minimized by reducing the temperature in the spray drying process which could be 
achieved through minimizing the probability of thermal degradation.

2.13.4 Water Solubility Index

The range of water solubility index (WSI) of Gau fruit aril powder was 36.91–38.25% 
which was not influenced by different drying conditions and maltodextrin 
concentration [193]. Mishra et al., [231] concluded that the drying temperature did not 
show any significant effect on WSI of the spray dried amla juice powder at 5% 
probability level.

2.13.5 Micro Structural Analysis

The morphology of spray dried cactus pear microcapsules was examined by 
scanning electron microscope. It was showed that microcapsules were irregularly 
spherical in shape with an extensively dented surface. The formation of the dented 
surfaces of the spray dried particles was attributed to the shrinkage of the particles 
during the drying process [232].

Norziah et al., [233] and Kurozawa et al., [234] reported that scanning electron 
microscopy in fish oil encapsulated powders and meat protein hydrolysate powder
showed some spherical, smooth and many particles presented a spherical shape with a shriveled surface when increasing maltodextrin. This may be due to rapid particle shrinkage during the early stage of the drying process [235].

Villacrez et al., [236] confirmed that the nozzle diameter has no significant influence on the morphology of encapsulated andes berry (*Rubus glaucus* Benth.) extract microcapsules. Roughness observed on the surface of the spheroidal particles may be from a hollow particle structure with thin floppy walls.

### 2.13.6 Applications of Microencapsules in Food System

Rocha et al., [237] incorporated lycopene microcapsules with modified food starch - capsul as encapsulating agent in cake preparation which was compared with free-form lycopene and a standard cake (without addition of lycopene). The colour values were examined using a colorimeter (Hunter Lab Color Quest II–VA, United States) and the reading was observed with the D65 illuminant, 10° opening for angle of vision at RSIN mode. The colour values were observed to be significantly different from each other. The cake made with microcapsules was more pigmented (“a” value - 26.59±0.61) than the standard cake (“a” value - 1.70±0.21), but the one made with free-form lycopene (“a” value - 33.65±0.46) was the product with the most pigmentation. It was observed that the microcapsules were able to release pigment and color the studied food system in a homogenous manner, which reveals efficacy of encapsulant functioning in cake pigmentation.

### 2.14 COST ECONOMICS

The cost of operation was determined by estimating the fixed cost and variable cost. The fixed cost and capital recovery factor were estimated as described by Rajkumar [238]. The variable costs, such as wages, electricity charges, cost of repairs and maintenance, cost of raw materials, *etc.*, were calculated based on the data collected during the operation of the dryer.