Chapter 10
Spray Drying of Bioactives

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Abstract Spray drying is a common unit operation for converting solids from liquid materials into powders for preservation, ease of storage, transport and handling, and economic considerations. Although most often considered as a dehydration process, spray drying can also be effective as an encapsulation method when it is used for complexing a core material with a protective matrix, which is ideally inert to the core material being encapsulated. Unlike other encapsulation techniques, it offers the unique advantage of producing microcapsules in a cost-effective one-step continuous process. This chapter describes the principles and processing techniques of spray drying for encapsulation of food bioactives, including probiotics, polyphenols, enzymes and peptides, vitamins, and essential fatty acids. The storage stability of spray dried bioactives and challenges from both a research and industrial perspective are also briefly discussed.

Keywords Spray drying · Encapsulation · Bioactive compounds · Stability

10.1 Introduction

Over the past decade, there has been an increasing awareness regarding the importance of maintaining overall health and wellness by adopting a positive lifestyle and consuming a healthy diet. Health-promoting foods are those considered to be beneficial to health in ways that go beyond a normal healthy diet required for human nutrition, and the term may also refer to functional foods that are designed to address specific health concerns or disease prevention (Roberfroid

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2000). Food components that contribute to these health benefits are bioactive compounds typically including vitamins, ω-3 fatty acids, peptides, and phytochemicals. However, most bioactive compounds are extra-nutritional constituents that occur only in small quantities in foods (Kris-Etherton et al. 2002), and their stability and functionality are highly dependent on processing and storage conditions. Research on the extraction and isolation of bioactives and on the development of bioactive supplements for human health is a hot topic in both academic and industrial communities (Vinatoru 2001; Kitts and Weiler 2003; Puri et al. 2012).

One practical means of preserving stability and bioactivity of these bioactive compounds is through the selection of a suitable processing method and storage of the compounds in a dry glassy state to reduce deterioration (Ludescher et al. 2001). Spray drying is a well-established method for transforming solids from liquid materials into solid dry powders. With the advantages of economical, flexible, continuous operation, and resulting powder with good flowability, this technique has been widely applied in chemical, food, biochemical, and pharmaceutical industries (Broadhead et al. 1992; Bhandari et al. 2008). Because the drying time is very short (generally 15–30 s) for the passage of the sprayed particle, and intense evaporation occurs at the surface to maintain the droplet at an almost cool temperature until a dry state is reached (Fogler and Kleinschmidt 1938; Gharsallaoui et al. 2007), spray drying is commonly used for drying of heat-sensitive materials containing flavors, enzymes, and probiotics (Ré 1998; Jafari et al. 2008b). In addition, as the spray dried material has a matrix structure that can entrap active (core) materials, spray drying is also used to encapsulate food ingredients (Gharsallaoui et al. 2007) and food bioactives (Fang and Bhandari 2010), which impart numerous benefits to the encapsulated compounds including significantly improved stability, bioactivity, and targeted release (Augustin and Hemar 2009; de Vos et al. 2010). For example, spray drying of milk can be considered as an encapsulation process where milk fat is the core material that is protected against oxidation by a wall material consisting of a mix of lactose and milk proteins (Gharsallaoui et al. 2007). It is therefore understandable that an encapsulation effect is desirable for the spray drying of bioactive compounds. This chapter will provide an overview of the spray drying of food solids with bioactives, and the stability of the powders will be discussed.

10.2 The Spray Drying Process

The concept of spray drying was first reported in 1872 in a patent by Percy (1872). The technique was introduced for commercial purposes in 1903, and was fully established on a large-scale basis in the early 1950s (Masters 1991; Bhandari et al. 2008). The basic process of spray drying is illustrated in Fig. 10.1. Briefly, it involves feeding a prepared liquid into a spray dryer, and atomization with a nozzle or rotating disc in a chamber supplied with dry hot air. The droplet comes into contact with the hot air in the chamber, causing evaporation of the solvent, and the dried particles are then separated and the fines removed from the humid air using a cyclone or bag filter (Ré

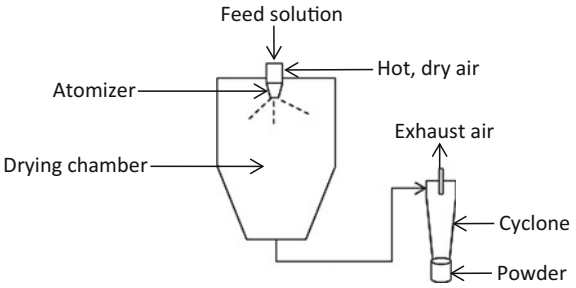

Fig. 10.1 Schematic of typical spray drying process

1998; Gibbs, et al. 1999; Zuidam and Shimoni 2010; Fang and Bhandari 2012a). Although less common, spray drying of ingredients from an organic solution such as acetone or ethanol may be performed if the materials are not water-soluble and are very heat-sensitive, requiring low-temperature drying. The spray drying process used for encapsulation of food bioactives is similar to that of dehydration, except that the feed solution preparation step may involve dissolving, emulsifying, or dispersing the core material in an aqueous medium with the selected wall material. The spray dried particles generally form a spherical matrix structure, but particle size may vary from very fine (10–50 μm) to large (2–3 mm) if spray drying is combined with an agglomeration process (Gharsallaoui et al. 2007). Particle sizes ranging from 300 nm to 5 μm were recently achieved with a BÜCHI Nano Spray Dryer B-90, which uses a vibrating mesh technology to generate fine droplets (Schmid et al. 2011).

10.2.1 Preparation of Feed Solution

Methods for preparing feed solutions are dependent on the nature of the material being spray dried and the end use of the powder. Water-soluble materials can be dissolved in water, but an oil-in-water emulsion should be prepared for oils or oil-soluble materials, and a solid/liquid suspension can be formed by dispersing a solid or particulate material (e.g. probiotics) in a solvent (Fang and Bhandari 2012a). It should be noted that if spray drying materials are sugar-rich products such as fruit/vegetable juice or honey, high molecular weight additives (starches, maltodextrins) or surfactants (proteins) may be added to overcome the problems of stickiness and agglomeration in the drying chamber (Bhandari et al. 2008; Fang and Bhandari 2012b; Fang et al. 2013;). For encapsulation, a wall material is selected and incorporated into the feed solution/emulsion/dispersion to protect the core materials. The wall material should have specific properties, including good film-forming and emulsifying ability, high solubility in water, low viscosity with high solids levels, low hygroscopicity, protection and controllable release of the core material, a stable and low-cost supply, and bland taste (Ré 1998; Desai and Park 2005; Augustin and Hemar 2009). Examples of two commonly used wall materials, carbohydrates and proteins, are presented in Table 10.1, along with the properties that they provide (Fang and Bhandari 2012a).

Table 10.1 Some commonly used wall materials and their properties for spray drying encapsulation (Fang and Bhandari 2012a, with permission)

Wall material	Example	Encapsulation-related properties
Carbohydrates	Hydrolyzed starches: corn syrup solids, maltodextrins	Very good oxygen barrier; low viscosity at high solids; no/limited emulsion stabilization; low cost
	Modified starches: acetylated starch, monostarch phosphate	Good emulsion stabilization; varying quality; constricted usage due to regulatory situation; low cost
	Cyclodextrins: α-, β-, γ-cyclodextrins	Good inclusion of volatiles; excellent oxygen barrier; relatively expensive
	Gums: agar, acacia (arabic), xanthan, alginates	Good emulsions; very good retention of volatiles; varying quality; price depends on availability; sometimes impurities
Proteins	Milk proteins: whey proteins, caseinates, skim milk powders	Good emulsions; properties dependent on other factors such as pH and ionic strength; allergenic potential; relatively expensive
	Other proteins: soy protein, egg protein, gelatin	
Other biopolymers	Soluble soy polysaccharides, chitosan, Maillard reaction products, modified celluloses	Varied properties; may provide additional benefit to the stability of actives

10.2.2 Atomization

Atomization is one of the most important processing steps during spray drying. The goal of this stage is to create a maximum heat transfer surface area between the liquid droplets and the dry medium in order to optimize heat and mass transfer. Liquid atomization in small droplets can be generated by atomizers. Commonly used types include rotary wheel/disc (using centrifugal energy), pressure nozzles (using pressure energy), dual-fluid-spray nozzles (using pressure and gas energy), and sonic nozzles (using sonic energy) (Masters 1991; Bhandari et al. 2008). Atomizers can be used either as an individual device or in groups, but must be able to work effectively and reliably under harsh conditions. The selection and operation of the atomizer is critical in spray drying, because drying efficiency, particle size, size distribution, and powder collection efficiency are all dependent on atomizer performance. The choice of atomizer also depends on the nature and viscosity of the feed solution and the desired characteristics of the dried powder. For example, the use of a pressure nozzle will enable the formation of finer droplets with higher energy input. At a constant energy level, the size of the formed particles increases with increasing feed rate. However, particle size increases when both the viscosity and surface tension of the initial liquid are high (Gharsallaoui et al. 2007; Bhandari et al. 2008). Special consideration for atomizer selection is required in spray drying encapsulation, especially when the feed solutions are solid-in-oil-in-water (s/o/w) emulsions or emulsions with high viscosity, because blockage of the atomizer can

occur with solid core material due to large particle size or viscosity that is too high (Fang and Bhandari 2012a). It is important, therefore, to control the solid core particle size and viscosity of the liquid emulsion before spray drying and to select an appropriate atomizer for a specific feed solution.

10.2.3 Drying Process

The drying process is initiated upon contact of the atomized liquid droplets with hot air in the drying chamber. Based on atomizer emplacement in relation to the inlet of hot air, spray drying can be classified as co-current or counter-current drying. In co-current drying, the hot air and the atomized droplets move in the same direction in the drying chamber, whereas with a counter-current drying design, the flow of hot air and the droplets move in opposite directions. During a co-current drying process, the mean residence time of the particles is small, and the dried particles do not have to pass through the high-temperature zone; therefore, it is suitable for drying thermo-sensitive materials such as bioactive compounds (Bhandari et al. 2008). Typically, the hot air inlet temperature ranges from 150 to 220 °C, and evaporation occurs instantaneously. The dry powders are thus exposed to moderate temperatures ($\sim$50–80 °C), which limits their thermal degradation (Fleming 1921; Gharsallaoui et al. 2007). For spray drying encapsulation, the short time exposure to keep the core temperature below 40 °C helps to prevent damage to the product (Dubernet and Benoit 1986). On the other hand, counter-current spray drying also has advantages. Because the dry product will be exposed to high temperatures during the drying process, the powders will generally have low final moisture content, which is of benefit to the shelf life of the dried product. Another advantage of the counter-current process is that it is considered more economical in terms of energy savings (Gharsallaoui et al. 2007).

At the time of droplet–hot air contact, heat transfer from the air towards the product occurs as a result of the temperature difference, and the transfer of water is carried out in the opposite direction due to the vapor pressure difference. Therefore, a balance of temperature and vapor partial pressure is established between the liquid and gas phases, and the loss of moisture from the droplet is controlled by the gas phase resistance. This is called the constant-rate or constant-activity period, as the water activity at the droplet surface remains nearly constant (Ré 1998). If the partial pressure of water vapor in the bulk air has not built up to a substantial value, the water vapor partial pressure driving force for mass transfer in the gas boundary layer surrounding the droplet remains constant, and the evaporation rate per unit area of droplet surface is constant. As heat continues to transfer by convection from the drying air to the surface of the droplet, water continues to evaporate from the surface. The water is lost in proportion to the heat gained, and the droplet temperature is the wet-bulb temperature of the drying air. As drying continues, a water concentration gradient is built up within the droplet, the water activity at the surface decreases, and the surface dries out. This brings about the falling-rate period, where drying is rate-limited by moisture transport within droplets. The relative lengths of the constant-rate and the falling-rate periods vary according to the conditions in the spray dryer and the material being dried. Once a

dry skin has been formed, the droplet temperature starts to increase from the wet-bulb to the air temperature. At temperatures reaching or exceeding the boiling point of water, substantial internal voids and particle inflation tend to occur. When the particle is sufficiently dry, the final shape sets into place, and any final evaporation of water occurs, thus completing the transformation of droplets into dry particles.

10.2.4 Recovery of the Spray Dried Powder

The dried powders are delivered from the drying chamber to a powder separator by the drying air flow. For modern multi-stage spray dryers, the air flow from the drying chamber typically contains about 10–50 % of the total powder, depending on the material and spray dryer type and operating conditions (Pisecky 1997). It is important to recover the powder from the exhaust flow not only for cost purposes, but also to clean the air in order to minimize pollution. In general, particles are recovered by incorporating one or more of the cyclone separators, bag filters, or wet scrubbers (Bhandari et al. 2008). With the use of a cyclone, particles hit the cyclone wall due to the centrifugal force and then drop down due to gravity, while the clean air moves up from the bottom to the top of the cyclone as a result of differential pressures created between the radial and axial directions. Bag filters are used in particular for the removal of the finest powders. With the wet scrubber, the exhaust air is injected at very high velocities through a venturi inlet. At the same time, a solvent fluid (normally water) is sprayed in such a way that the gas stream and water spray make intimate contact. The fine particles are dissolved in the water, and the clean air then escapes from the scrubber.

After separation, the obtained powder is composed of particles originating from spherical droplets with some degree of shrinkage. The particles can be compact or hollow, depending on the composition, drying temperature and water and gas content of the droplet (Bimbenet et al. 2002). A fluidized bed is sometimes integrated into the spray dryer to reduce drying cost, better control the particle size, or produce powders with very low water content (Fang and Bhandari 2012a). In addition, an agglomeration process may be utilized after spray drying to improve the rehydration ability of the dried particles.

10.3 Applications of Spray Drying for Drying of Bioactives

Bioactive compounds *can be defined as natural essential and non-essential compounds (e.g. vitamins or polyphenols) in the food chain that have an effect on human health* (Biesalski et al. 2009). *Because of their sensitivity to adverse environments such as high temperature, light, and oxygen, it is important to select a suitable processing method to preserve their stability, and therefore their bioactivity. Spray drying is a widely accepted and economical method for drying of heat-sensitive materials* (Bhandari et al. 2008)*, including bioactive compounds.*

10.3.1 Probiotics

The word 'probiotics' is derived from the Latin 'pro' and the Greek 'biotic', meaning 'for life', and was defined by Fuller (1992) *as 'a live microbial feed supplement which beneficially affects the host animal by improving its intestinal microbial balance'*. Probiotics can survive gastric, bile, and pancreatic secretions, attach to epithelial cells, and colonize the human intestine. Microbes from many different genera are currently used as probiotics, among which the most common strains are *Lactobacillus* spp. (*L. acidophilus, L. casei, L. johnsonii, L. plantarum*), *Bifidobacterium* spp. (*B. bifidum, B. animalis, B. breve*), and yeasts (*Saccharomyces boulardii*) (Alvarez-Olmos and Oberhelman 2001). Probiotics have been reported to provide a variety of beneficial health effects and are used in a number of therapeutic applications, including maintenance of normal gastrointestinal tract microflora, improvement of constipation, treatment of diarrhea, enhancement of the immune system, alleviation of lactose intolerance, reduction of allergy risk in infancy, reduction of serum cholesterol levels, anticarcinogenic activity, and improved nutritional value of foods (Kailasapathy and Chin 2000; Mattila-Sandholm et al. 2002; Touhy et al. 2003). In the food industry, a large number of probiotic products are manufactured in the form of milk, drinking and frozen yogurts, probiotic cheeses, ice creams, dairy spreads, and fermented soy products (Manojlović et al. 2010).

For spray drying of probiotics, the low survival rate of the microorganism during drying and low stability upon storage are the major factors limiting its commercialization (Manojlović et al. 2010). The damage to the cell wall, cytoplasmic membrane, ribosome, and DNA during the heating process may be the main reasons for the loss of viability (Abee and Wouters 1999). As discussed by Peighambardoust et al. (2011), the viability of probiotics during spray drying is dependent on a wide range of factors, including biological parameters (species, growth media, growth phase, intrinsic stress tolerance), processing parameters (inlet and outlet temperature, drying time, nozzle pressure), product parameters (carrier medium, concentration), pretreatments (stress response, protective substances), and post-drying conditions (rehydration, packaging, and storage).

The survival of probiotics during spray drying depends on the air temperature, types and strains of probiotics, and type of protective agent used. Moderate spray drying temperature conditions (air inlet temperature <160 °C) are usually selected for drying probiotics, as a very high temperature is detrimental to the viability of the probiotic bacteria (Anal and Singh 2007; Chavez and Ledeboer 2007). In one study, a lower inlet air temperature of 100 °C and outlet air temperature of 45 °C was found to obtain higher viability for spray drying encapsulation of *Bifidobacterium* sp. than conditions of higher inlet (>120 °C) and outlet (>60 °C) temperatures (O'Riordan et al. 2001). Other researchers have reported that the survival of probiotics declined with increasing inlet temperature (Mauriello et al. 1999; Gardiner et al. 2000), whereas outlet temperature was even more crucial to their survival (Ananta et al. 2005; Chavez and Ledeboer 2007; To and Etzel 1997). However,

care should be taken to ensure that the powder obtained at lower outlet temperatures has been dried sufficiently, because higher moisture content and higher water activity is obtained after low inlet/outlet temperature drying that will affect storage stability. The survival of probiotics is optimal at low water activity (<0.25) and low temperatures during storage, and a nitrogen- or vacuum-sealed package with a proper barrier function is recommended for storage of spray dried probiotics (Manojlović et al. 2010).

The viability of distinct species of a given genus and the growth phase differ under the same drying or storage conditions (Simpson et al. 2005). One group reported that the survival rate after spray drying was greatest for *Streptococcus thermophilus*, followed by *Lactobacillus paracasei* ssp. *paracasei*, with *L. lactis* ssp. *cremoris* found to be the least heat-tolerant microorganism (To and Etzel 1997). Lactic acid bacteria harvested at the stationary phase showed enhanced viability after spray drying (Corcoran et al. 2004), mainly due to the depletion of nutrients and glucose starvation in bacterial cells in the stationary growth phase. The cells demonstrated resistance to many environmental stresses, including osmotic and heat stress (Van de Guchte et al. 2002). Some studies have reported that the type of nozzle can influence the survival of the spray dried probiotics. An ultrasonic atomization nozzle provided smaller and more uniform particles compared to the traditional centrifugal and stationary dual-fluid spray nozzles, and resulted in smaller numbers of microorganism after drying (Al and Al 2009). The use of relatively low nozzle pressure has also been suggested to avoid high shear stress on the probiotics. For example, higher viability of *Lactobacillus acidophilus* was achieved after spray drying when the atomization pressure was reduced from 100 to 50 kPa (Riveros et al. 2009), and increased survival was reported for *Lactobacillus bulgaricus* when a spray pressure of 100 kPa instead of 200 kPa was used (Lievense and van Riet 1994).

The addition of prebiotics can also improve the survival of probiotics during spray drying and storage. It was reported that adding prebiotic oligosaccharides to whey protein isolate, pectin, and carboxymethylcellulose improved the survival of spray dried *L. acidophilus* (Vila-Garcia et al. 2010). Spray drying of the prebiotics inulin and oligofructose were reported to increase the storage survival rate of *Bifidobacterium* BB-12 (Fritzen-Freire et al. 2011). These prebiotics act as protective agents and sometimes also as carrier materials that reduce heat and shear stresses during drying and storage. Other protective agents include simple or complex components such as sugars (e.g. glucose, fructose, lactose, mannose, sucrose, sorbitol, adonitol, trehalose), ascorbic acid, skim milk, acacia gum, monosodium glutamate, and starch (Peighambardoust et al. 2011). Typically, these protective agents/carrier materials are combined with microorganisms at a concentration of about 10–20 % (w/w) (Lian et al. 2002; Ananta et al. 2005). Feed solutions with solid content that is too high (e.g. 40 %) produce larger particle sizes with longer drying times, and consequently greater thermal inactivation and less viability of the bacterial cells (Santivarangkna et al. 2007).

10.3.2 Polyphenols

Polyphenols are a group of plant secondary metabolites with a basic structure of several hydroxyl groups on aromatic rings (Manach et al. 2004). The most common and widely distributed group of polyphenols is flavonoids. Flavonoids are diphenylpropanes (C6-C3-C6) and consist of two aromatic rings linked through three carbons that usually form an oxygenated heterocycle (Fig. 10.2). Other common polyphenols include phenolic acids, stilbenes, and lignans, as presented in Fig. 10.2 (Manach et al. 2004). Interest in polyphenols has increased because of the recognition of their antioxidant properties, their great abundance in the human diet, and their likely role in the prevention of various diseases associated with oxidative stress, such as cancer and cardiovascular and neurodegenerative diseases (Scalbert et al. 2005). In addition to antioxidant activity, polyphenols possess potential anti-inflammatory, antibacterial, and antiviral functions, suggesting a wide range of health benefits to humans (Surh 2003; Manach et al. 2004; Scalbert et al. 2005). However, most polyphenols are highly unstable under adverse environmental conditions such as unfavorable temperatures, light, pH, moisture, enzymes, and oxygen, and are therefore susceptible to degradative reactions in the course of food processing and storage or in the gastrointestinal tract (Bell 2001). Thus, the delivery of polyphenols requires protective properties that can maintain the active molecular form until the time of consumption, and that deliver this form to the physiological target within the organism. One natural characteristic of polyphenols is that they may have an unpleasant taste, such as astringency and bitterness, which needs to be masked before they are used in food or nutraceutical products (Haslam and Lilley 1988).

Spray drying has been used successfully for encapsulation of polyphenols and for maintaining their stability and antioxidant activity, as a number of studies have reported high polyphenol/antioxidant activity recovery (Table 10.2). Saéna et al. (2009) and Robert et al. (2010) found extremely high polyphenol recovery, exceeding 100 %, which may have been due to the hydrolysis of polyphenol conjugates during the preparation of the samples or during the drying process.

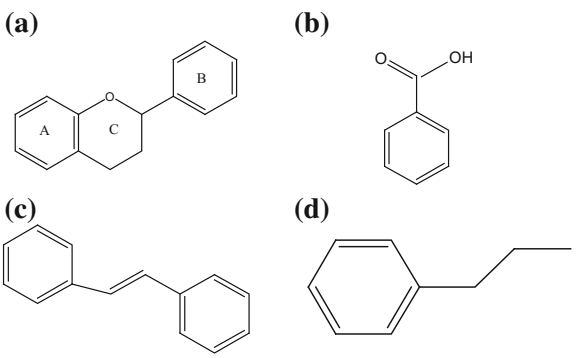

Fig. 10.2 Basic structures of polyphenols: **a** flavonoids, **b** phenolic acids, **c** stilbenes, and **d** lignans

However, high polyphenol degradation (65–68 %) was also reported by Georgetti et al. (2008), although the reasons were not mentioned, and the definition and calculation of polyphenol degradation in the study was unclear. Because most polyphenols are water-soluble compounds, an acceptable level of water solubility must also be present in the wall materials used in spray drying encapsulation, such as starch, maltodextrin, gum arabic, sodium caseinate, whey proteins, or their combinations (Table 10.2). The spray dried particles generally form a matrix structure of wall and core materials and have a spherical morphology.

In spray drying encapsulation of polyphenols, the air inlet temperature plays a critical role in polyphenol degradation; generally, the higher the temperature, the lower the polyphenol stability. In a study investigating spray drying of soybean extracts, an increase in the inlet gas temperature led to a product with a lower polyphenol concentration (Georgetti et al. 2008). In addition, greater loss of anthocyanin was observed with a higher (>180 °C) versus lower (160 °C) air inlet temperature (Ersus and Yurdagel 2007). The drying temperature is also affected by the encapsulation wall materials. For example, for spray drying encapsulation of cactus pear pulp and ethanol extract, an inlet temperature of 140 °C was used when

Table 10.2 Recent reports on spray drying encapsulation of polyphenols

Core materials (polyphenols)	Wall materials	Recovery	References
Bayberry juice (phenolic acids and flavonoids)	Maltodextrin and whey protein isolate	Polyphenols > 94 %	Fang and Bhandari (2011, 2012b)
Black carrot extracts (anthocyanins)	Maltodextrins	Not provided	Ersus and Yurdagel (2007)
Cactus pear pulp and extract (flavonoids and betalains)	Maltodextrin and inulin	Polyphenols > 100 %, betalains 62–100 %	Saéna et al. (2009)
Grape seed extract, apple polyphenol extract and olive leaf extract	Sodium caseinate-soy lecithin	Antioxidant activity 60–80 %	Kosaraju et al. (2008)
Hibiscus sabdariffa L. extract (anthocyanins)	Citrus fruit fiber	Not provided	Chiou and Langrish (2007)
Olive leaf extract	Chitosan	"No inactivation" of polyphenols	Kosaraju et al. (2006)
Pomegranate juice and ethanol extract (anthocyanins)	Maltodextrin and soybean protein isolates	96–121 %	Robert et al. (2010)
Procyanidins	Maltodextrin and gum arabic	Procyanidins > 89 %	Zhang et al. (2007)
Soybean extract	Colloidal silicon dioxide, maltodextrin, and starch	32–36 %	Georgetti et al. (2008)

maltodextrin was used as wall material, while a temperature of 120 °C was applied with inulin as wall material (Saéna et al. 2009). The wall material also affects the stability of polyphenols. Georgetti et al. (2008) reported that the degradation of polyphenol after spray drying was reduced in soybean extract when colloidal silicon dioxide (Tixosil 333) was added as a drying aid to the wall materials of maltodextrin and starch. Therefore, optimization of spray drying conditions, especially drying temperature and wall material, is critical to the success of polyphenol encapsulation.

10.3.3 Enzymes and Peptides

Enzymes and peptides are protein-based molecules that perform a variety of functions in organisms. Enzymes are capable of catalyzing one or more specific types of chemical or biochemical reactions, and have been used in a wide array of industrial applications, including cosmetics, textiles, feed, and food industries, and even for the production of fuel alcohol and organic synthesis (Kirk et al. 2002). Peptides consist of short amino acid sequences that have less intricate functionality than proteins, and are most often used in the pharmaceutical, biomedical (de la Rica and Matsui 2010), and food industries (Molina Ortiz et al. 2009; da Silva Malheiros et al. 2010). To maintain their viability during an extended shelf life and to protect their stability and bioactivity, enzymes are often encapsulated or immobilized for commercial applications (Gibbs et al. 1999). The encapsulation of peptides is performed mainly to attenuate their bitter taste (Molina Ortiz et al. 2009; Favaro-Trindade et al. 2010) and to protect their bioactivity (de la Rica and Matsui 2010). Spray dried enzymes include trypsin, amylases, proteases, glucose oxidase, pectinases, glucose isomerase, lactase, and pepsin (Bhandari et al. 2008), while spray dried peptides include casein hydrolysate (Molina Ortiz et al. 2009; Favaro-Trindade et al. 2010), nisin (de la Rica and Matsui 2010), capreomycin (Schoubben et al. 2010), and chicken meat protein hydrolysate (Kurozawa et al. 2011).

A key consideration in utilizing spray drying encapsulation of enzymes and peptides is how well these thermally labile materials resist heat denaturation by hot air. At the early stage of drying, when the droplet surface remains moisture-saturated (100 % relative humidity), its temperature is maintained at the wet-bulb temperature, which is significantly lower than the hot air temperature. As drying continues, the droplet temperature begins to rise as the diffusion of water to the droplet surface can no longer maintain 100 % moisture. At this stage, the protein is primarily in a solid state, and the surrounding air temperature decreases significantly due to moisture uptake (Ameri and Maa 2006). Thus, thermal denaturation is not typically observed in spray drying. In practice, the use of a lower inlet air temperature is advisable to reduce the potential thermal stress to the protein. However, protein denaturation typically occurs during dehydration, so the enzymes and peptides must be dried in the presence of wall materials and/or protective

materials such as lactose, sucrose, mannitol, gums, maltodextrins, or cyclodextrins (Daeman and van der Stage 1982). The types of protective agents added to wall materials can affect the activity of enzymes; for example, sucrose was reported to be more effective than trehalose in stabilizing lysozyme (Liao et al. 2002), but trehalose was found to be superior to mannitol, sucrose, and arginine hydrochloride as a stabilizer for spray drying of β-galactosidase (Broadhead et al. 1994). The authors suggest that sugars can protect lysozyme against dehydration stress by hydrogen bonding between the sugar and protein molecules.

Two other possible sources of stress during spray drying of enzymes and peptides are atomization and the air–water interface (Ameri and Maa 2006). Mathematical modeling estimated that the shear rate imparted on the protein from atomization is in the range of 10^4–10^5 s^{-1}, and proteins can sustain shear rates as high as 10^5 s^{-1} (Maa and Hsu 1996). Therefore, shear stress of this magnitude does not impart a significant stress to the enzymes and peptides. However, when shear stress is combined with air–water interface, it may cause significant aggregation for air–water interface-sensitive proteins such as recombinant human growth hormone (rhGH), bovine serum albumin (BSA), and lactate dehydrogenase (LDH) (Faldt and Berganstahl 1994; Maa and Hsu 1996; Adler and Lee 1999). Hence, it is suggested that comprehensive factors should be considered in spray drying of enzymes and peptides, such as relatively lower inlet and outlet air temperatures (typically 120–140 °C and 50–65 °C, respectively), suitable protective materials and total solids content (around 10–20 %), and low shear during atomization (Broadhead et al. 1992).

Most enzymes and peptides are amphiphilic compounds, and thus are susceptible to adsorption at the air–water droplet interface, where unusual surface energies may cause the protein to unfold, exposing hydrophobic regions to hot air (Ameri and Maa 2006). The unfolded protein may then undergo aggregation by the interaction of the exposed hydrophobic regions with other unfolded molecules until precipitation occurs. The enzymes and peptides on the powder particle surface may experience the most severe drying and shear-induced stress, and may degrade faster during long-term storage, primarily due to the partial loss of their secondary structure caused by temperature, moisture, and other environmental stresses related to drying. To mitigate surface denaturation, three options have been recommended for minimizing rhGH aggregation: the addition of a surfactant to prevent the formation of insoluble aggregates, the addition of divalent zinc ions to prevent the formation of soluble aggregates, and increasing the rhGH concentration in the liquid feed (Maa et al. 1998). The addition of a surfactant such as polysorbate 80 to the lactate dehydrogenase (Adler and Lee 1999) and Tween 80 to trypsin (Millqvist-Fureby et al. 1999) wall materials was reported to prevent enzyme inactivation during spray drying. The addition of surfactant is thought to be helpful for reducing the enzyme concentration on the particle surface, as the surfactant molecules are preferentially adsorbed at the air–liquid interface of the droplets, thus expelling proteins from the surface. The loss of enzyme and peptide activity can be estimated by incorporating the inactivation kinetics model with the drying kinetics model, because the inactivation rate is temperature/time- and moisture

content/time-dependent. Some existing inactivation kinetics models and relevant issues within the context of spray drying of food materials with bioactive compounds were recently reviewed by Chen and Patel (2007), and an appraisal showing the effect of surface and center water concentrations on the inactivation rate was proposed.

10.3.4 Vitamins

Vitamins are minor but essential constituents of foods. They are required for the normal growth, maintenance, and functioning of the human body, and insufficient supply can lead to many deficiency diseases, such as scurvy, pellagra, ariboflavinosis, dermatitis, and enteritis (Belitz et al. 2009). Hence, vitamin preservation during the processing and storage of foods is of critical importance. The use of multivitamin supplements has been reported to reduce the risk of certain diet-related disorders (Pocobelli et al. 2009).

Vitamins are usually divided into two general classes: fat-soluble vitamins, such as A (retinol and carotenoids), D (calciferol), E (α-tocopherol), and K (phytomenadione); and water-soluble vitamins, such as B1 (thiamine), B2 (riboflavin), B6 (pyridoxine), niacin, pantothenic acid, biotin, folic acid, B12 (cyanocobalamin), and C (L-ascorbic acid). However, vitamins are completely or partially deactivated or damaged during the cooking and processing of foods. The average loss of vitamins through processing/preservation of vegetables and fruits varies from 10 to 60 %, depending on the fruit and vegetable varieties and the techniques employed, with vitamin C, folic acid, and vitamin B6 generally less stable during high-temperature processing than vitamins A, B1, and B2, and niacin (Belitz et al. 2009). Micro- and nano-encapsulation, including spray drying, are good techniques for preserving vitamins (Murugesan and Orsat 2012). Due to their processing stability and commercial value, vitamins C and A (mainly carotenoids) are those most often used for spray drying encapsulation.

Microencapsulated ascorbic acid microparticles have been reported to prevent ascorbic acid color change, slow its core release rate, and generally mask its acidic taste (Uddin et al. 2001). Under spray drying inlet and outlet temperature parameters of 200–300 °C and 70–95 °C, respectively, the loss of ascorbic acid during encapsulation was found to be minimal (less than 2 %), and was much lower than with other encapsulation methods such as thermal phase separation, melt dispersion, and solvent evaporation (Uddin et al. 2001). The selection of a suitable encapsulation wall material is important for ensuring delivery of the ascorbic acid to the target organ for controlled release. For example, methacrylate copolymer of Eudragit® L is a pH-dependent enteric polymer composed of methacrylic acid–methacrylic acid methyl ester copolymers soluble from pH 6, which suggests that it is insoluble in the mouth and in the stomach, and soluble in the duodenum (pH around 6) (Weiss et al., 1993). Since the pH in the colon is around 7.5, researchers used Eudragit® L as a wall material in spray drying encapsulation for the delivery of

ascorbic acid to the lower part of the intestine and the jejunum and ileum (Esposito et al. 2002). The encapsulation of ascorbic acid with methacrylate showed very high efficiency, around 98–100 %, with good morphology and size distribution, although this was unable to slow the release of the drug with respect to the free form of ascorbic acid. In another study, in order to prepare drugs with sustained-release capability, vitamin C was encapsulated in tripolyphosphate (TPP)-cross-linked chitosan (TPP–chitosan) microspheres using spray drying (Desal and Park 2006). The authors found that an inlet temperature of 170 °C, liquid flow rate of 2 ml min^{-1}, and compressed air flow rate of 10 l min^{-1} provided optimal drying conditions, with encapsulation efficiency ranging from 45.7 to 68.7 %. The rate of vitamin C released from the TPP–chitosan microspheres decreased with increasing TPP solution volume and chitosan concentration (Desal and Park 2006). In these TPP–chitosan vitamin C microspheres, the molecular weight of chitosan also affected the encapsulation efficiency and controlled-release behavior, i.e. the encapsulation efficiency and release rate decreased with an increase in the chitosan molecular weight (Desal et al. 2006).

Spray drying is also a relatively effective method for encapsulation and preservation of carotenoids; one study reported that only 11 % degradation of carotene was observed in spray drying versus 14 % for drum drying (Desobry et al. 1997). Shu et al. (2006) also observed high encapsulation yield and efficiency, about 91 % and 82 %, respectively, in spray drying of lycopene. Because carotenoids are water insoluble, they were dissolved in oil phase to make an oil-in-water (O/W) and water-in-oil-in-water (W/O/W) multiple emulsions (Rodríguez-Huezo et al. 2004), or suspended in wall material solutions by homogenization (Desobry et al. 1997) before spray drying. Carotenoids of astaxanthin (Feldthusen et al. 2005), β-carotene (Loksuwan 2007), lutein (Reuscher et al. 2004), lycopene (Shu et al. 2006; Montenegro et al. 2007), and oleoresin (Rodríguez-Huezo et al. 2004; Santos et al. 2005), and samples containing carotenoids (Leuenberger et al. 2008) have been spray dried using encapsulating materials including gellan and mesquite gums, gum arabic, acacia gum, gelatin, skimmed milk, sodium caseinate, soy bean protein, trehalose, sucrose, lactose, maltodextrin, γ-cyclodextrin, pectin, cellulose, cellulose derivatives, modified starch, and modified polysaccharides, individually or in combination. Encapsulation yield and efficiency are dependent on the wall material formulation, core/wall material ratio, and homogenization pressure and temperature during the drying process (Shu et al. 2006).

10.3.5 Essential Fatty Acids

Essential fatty acids, such as ω-6 and ω-3 fatty acids, are those that humans and other animals require for good health but cannot synthesize in the body, and therefore must be ingested through the diet (Goodhart and Shils 1980). These fatty

acids, and especially the ω-3 fatty acids, are essential for normal growth and development, and may play an important role in the prevention and treatment of coronary artery disease, hypertension, diabetes, arthritis, other inflammatory and autoimmune disorders, and cancer (Simopoulos 1999). The ω-3 fatty acids in vegetable oils are primarily in the form of α-linolenic acid, with soybean oil and canola oil as important dietary sources (7–11 % of the total fat), whereas marine fish oils have a considerably higher content of ω-3 fatty acids (20–40 % of total fat), mostly in the form of eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA) (Alexander 1998). However, due to their highly unsaturated nature, the ω-3 fatty acids are very sensitive to lipid oxidation, which leads to a wide array of off-smells and off-tastes, varying from grass- and bean-like, to cardboard- and fish-like flavors (Beindorff and Zuidam 2010).

Encapsulation of essential fatty acids is a good method for slowing their oxidation, enhancing stability, controlling lipid-soluble flavor release, masking unpleasant taste and smell, and protecting dissolved substances against enzyme hydrolysis (Matsuno and Adachi 1993). However, the use of spray drying for encapsulation of fatty acids is a considerable challenge, as the spray drying process may induce oxidation due to the porous structure of the spray dried particles which allows high access of air to the oil, thus limiting the shelf life of the fatty acids (Hogan et al. 2003; Kolanowski et al. 2006, 2007). Therefore, the prevention of oxidation is of major concern for spray drying encapsulation of these highly unsaturated fatty acids. The extent of the protective effect is dependent on the nature of the wall materials, the fatty acids, and the encapsulation conditions (Matsuno and Adachi 1993). For example, the stability of fish oil was improved after spray drying in a polymer wall system of maltodextrin, modified starch, and whey protein concentrate (Jafari et al. 2008b), and good oxidation stability was reported for spray dried linoleic acid using whey protein concentrate as a wall material (Jimenez et al. 2004). In another study involving spray drying of linoleic acid, Minemoto et al. (2002) observed that wall material of gum arabic produced a better powder than maltodextrin in terms of encapsulation efficiency, stability, and oxidation resistance.

Lin et al. (1995) reported that the use of gelatin, sodium caseinate, and maltodextrin as wall materials for spray drying encapsulation was effective in improving the oxidative and thermal stability of crude squid oil, and that effectiveness was further enhanced by the addition of lecithin and carboxymethyl cellulose. Another study suggested that the stability of fish oil after spray drying was dependent on the interfacial composition and properties of both the oil–water interface in the parent emulsion and the surface composition of the drying droplets (Drusch et al. 2007). Emulsification of fish oil in a lecithin—chitosan multilayer system and the addition of EDTA were shown to have a positive effect on the stability of ω-3 fatty acids after spray drying (Shaw et al. 2007). The induction of simple coacervation by adding maltodextrin into the fish oil emulsion and hydroxypropyl methylcellulose (HPMC) solutions prior to spray drying was also found to increase oxidation stability (Wu et al. 2005). In addition, the esterification of polyunsaturated fatty acids with L-ascorbic acid and subsequent microencapsulation of the ester was demonstrated as a useful technology for slowing the

oxidation of linoleic acid (Jimenez et al. 2004). The particle size of the spray dried powder may also affect fatty acid stability; for example, it was noted that linoleic acid powder with larger particles was more stable during storage than that with smaller particles (Fang et al. 2005).

Other studies suggest that further improvement in the stability of encapsulated fatty acids may be achieved by conducting a Maillard reaction of wall materials of proteins (sodium caseinate, whey protein isolate, soy protein, or skim milk powder) with carbohydrates (glucose, dried glucose syrup, or oligosaccharides) at 60–100 °C for 30–60 min before spray drying, possibly because of changes in powder morphology and/or antioxidant effect of the Maillard products (Sanguansri and Augustin 2001; Augustin et al. 2006; Luff 2007). The addition of the antioxidants tocopherol or Trolox C to the oil prior to spray drying was also shown to improve the storage stability of spray dried fish oil (Hogan et al. 2003; Baik et al. 2004), although these compounds may act as pro-oxidants at high concentrations (Kolanowski et al. 2006). The use of trace metal chelation of citrem [citric acid esters of mono- and diglycerides of fatty acids] was found to improve the storage stability of the oil emulsion prior to spray drying but not after spray drying. The oil storage stability was significantly retarded by adding a combination of the antioxidants tocopherol, ascorbyl palmitate, and rosemary extract before the drying process (Serfert et al. 2009). In spray drying encapsulation of fatty acids, the fatty acid on the surface of particles may be released due to the rupture of the particle membranes that stabilizes them as an emulsion, which can result in high surface oil and free fat content in the dried powder. The released surface fat is prone to oxidation, as it is no longer encapsulated. Therefore, it is important to minimize the free fat content by selecting the correct drying conditions and formulation (Fang and Bhandari, 2012a).

10.4 Stability of Spray Dried Bioactives

Similar to other encapsulation processes, spray drying is not always efficient for the encapsulation of bioactives. Therefore, the un-encapsulated compounds, particularly those located on the powder particle surface, are prone to degradation during powder storage. Although spray drying conditions can be optimized to maximize encapsulation efficiency and retention of bioactive components upon drying, this may not necessarily result in good stability upon storage (Manojlović et al. 2010). Because of the small particle size and high surface area, spray dried carotenoid powder showed very fast degradation kinetics during storage (Desobry et al. 1997). Kolanowski et al. (2006, 2007) concluded that spray dried fish oil was even less stable against oxidation upon storage than fish oil, although this is not a common finding, and may be due to the drying conditions applied and/or materials used.

Generally, the storage stability of spray dried encapsulated bioactives is dependent on the formulation (e.g. wall and core materials, protective agents, emulsion type) before drying, operating conditions (e.g. inlet and outlet

temperature, nozzle type, air flow, feed speed) during drying, and packaging type and storage environment (light, humidity, and temperature) after the drying process. For example, the use of the relatively high-glass-transition-temperature (T_g) materials starch and β-cyclodextrin as encapsulating materials in spray drying delayed the degradation of ascorbic acid during storage and showed improved results over un-encapsulated ascorbic acid (Uddin et al. 2001). A formulation of lycopene-loaded O/W emulsion with gum arabic and sucrose mixture was demonstrated to slow the degradation of vitamins A and D3 during storage by 45 % compared with a formulation without lycopene (Montenegro et al. 2007). An air inlet temperature above 160 °C was observed to cause greater anthocyanin loss during spray drying and storage (Ersus and Yurdagel 2007). Interestingly, specific treatments of feed solutions before drying, such as the use of the Maillard reaction of protein–carbohydrate wall materials, has also been found to improve the storage stability of spray dried fish oil (Augustin et al. 2006) and ω-3 fatty acid (Luff 2007). The use of combined antioxidants (tocopherol, ascorbyl palmitate, and rosemary extract) before drying was reported to significantly improve the protective effects against autoxidation in spray dried fish oil during storage (Serfert et al. 2009). In addition, increasing the size of the emulsion and powder particles (e.g. 10–150 mm) may reduce the particle surface area and therefore improve storage stability (Soottitantawat et al. 2005).

Because most bioactive compounds are sensitive materials, the most common reactants must be avoided. These include adverse chemical, physical, and biological factors such as oxygen, moisture, light, elevated temperature, and microbial contamination (Morgan et al. 2006). The shelf life of spray dried fish oil (Kolanowski et al. 2007) and anaerobic probiotics of *Bifidobacterium* (Chávez and Ledeboer 2007) can be enhanced by packing them alone or in food powders under nitrogen or vacuum in metalized packaging material. The spray dried materials are believed to be more stable when stored at temperatures below their T_g, and lower storage temperature and water activity (a_w) generally lead to better stability, depending on the type of encapsulated core material. For example, the highest storage stability of spray dried bayberry polyphenols was found at 25 °C and with a_w of 0.33, and temperatures and a_w above this range caused faster polyphenol degradation and powder caking (Fig. 10.3) (Fang and Bhandari 2011). For spray dried enzymes and probiotics, one of the most important factors affecting their stability and activity during storage is residual moisture level, and the choice of optimal moisture content (on the order of 3–8 %) is a compromise between high survival rates immediately after drying (better survival with higher water content) and low rates of inactivation upon storage (better survival with lower water content, although not necessarily at 0 %) (Manojlović et al. 2010), such as moisture content of less than 5 % and a_w of less than 0.25 for storage of probiotics (Chávez and Ledeboer 2007). The addition of an appropriate amount of wall material is necessary to reduce the hygroscopicity and caking of encapsulated powders. After spray drying, further processing or coating (e.g. fluid bed coating and drying) may be necessary if a very high-quality

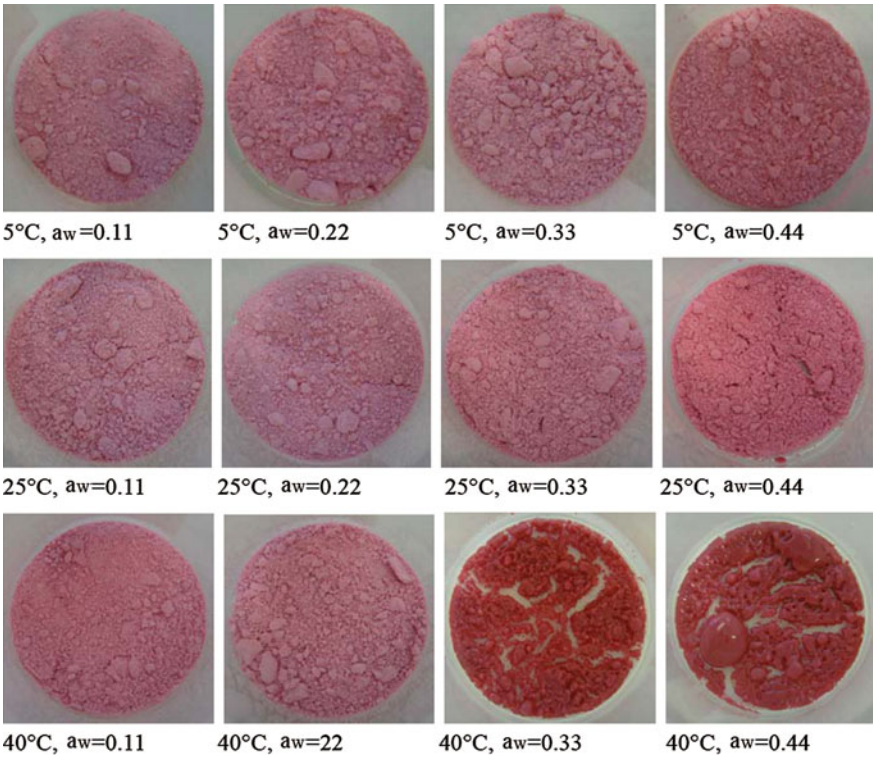

Fig. 10.3 Visual observations (digital camera photos) of spray dried bayberry polyphenol materials stored at different temperatures (5, 25, and 40 °C) and a_w (0.11–0.44) for 3 months (Fang and Bhandari 2011; with permission)

product is desired. Although it is possible to use very low temperatures for highly heat-sensitive bioactives, the process is not economically viable (Adhikari et al. 2009).

10.5 Conclusion and Future Research Opportunities

As discussed in other chapters of this book, a wide variety of techniques are available for the encapsulation and delivery of food bioactives, each with its own advantages and disadvantages. Spray drying represents a relatively simple continuous operation for producing food, nutraceutical, and pharmaceutical products with unique particle characteristics. Multiple factors must be optimized in spray drying encapsulation of bioactives, and this continues to be the subject of extensive research in order to manufacture high-value innovative products with extraordinary health benefits, in both the science and industrial sectors.

Research has shown that the spray drying step itself can be carried out successfully without difficulty, and can be optimized by trial-and-error procedures, but distinct improvements are needed in the choice of encapsulation materials to match the requirements of specific bioactive compounds (Gharsallaoui et al. 2007). The general requirements for wall materials are non-toxicity, low cost, bland taste, acceptable water solubility, good emulsion properties, and non-reactivity to core materials, as discussed in Sect. 2.1 and listed in Table 10.1. Because it is almost impossible for a single material to possess all of these properties, blends of multiple wall materials in one formulation are often applied. Therefore, numerous parameters within the formulation, including the type and concentration of wall polymer, core/wall ratio, type and concentration of heat/cold protectants, and type and concentration of emulsifier, may all contribute to the success of the encapsulation process (Fang and Bhandari 2012a). Each formulation is typically tailored to the specific type of core materials and specific spray drying conditions.

Innovation in spray drying technology is largely facilitated by combining existing novel technologies and the most recent knowledge of the processing and production sides (Bhandari et al. 2008). Several innovative designs and equipment modifications have been reported recently, including nano-spray drying (Lee et al. 2011), two-stage horizontal spray dryers (Huang and Mujumdar 2006), and spray-freeze drying (Maa et al. 1999). However, much work remains regarding product development, design quality, health and safety matters, environmental issues, and energy consumption. In addition, parameters and conditions in laboratory work should be fully tested and evaluated at pilot plants prior to large-scale production. Improvements in manufacturing technologies, new strategies for the stabilization of fragile bioactives, and the development of novel approaches will all contribute to the development of more efficient, economical, convenient, and simple spray drying techniques capable of satisfying demands across a wider range of industries for the manufacture of high-quality products.

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