

I Introduction

Microencapsulation is defined as “the technology of packaging solid, liquid, and gaseous materials in matrices or small capsules that release their contents at controlled rates over prolonged periods of time” [28;242;280]. The substance to be encapsulated is called “core”, while the microencapsulating agent surrounding the core is defined as “wall”. The core is also known as “active agent”, and the term “wall” is also referred to “matrix, coating material, or shell”. Microcapsules often have a diameter between 3 and 800 microns and contain 10 to 90 wt % core. The shell is designed to prevent diffusion of material from a microcapsule or into a microcapsule [282], to protect the core from deterioration, and to release it under the desired conditions [313]. Microencapsulation can suppress the volatility of flavor, odor and reactivity of food ingredients [118].

The first commercial application of microencapsulation technology began in the late 1930s and 1940s with the development of “carbonless paper” by the National Cash Register. Gelatine was used to coat a colourless dye precursor by a process known as coacervation. Using the same wall (gelatine) drugs were microencapsulated as early as 1931 [42]. Microcapsules are also used in applications of pharmaceuticals, pesticides and scented strips [251]. Nowadays, the encapsulation technology has been applied broadly in the food industry to microencapsulate sensitive food ingredients such as flavors, spices, vitamins, carotenoids, and omega-3 oils. The technology is aimed to protect sensitive ingredients from chemical degradation by blocking the direct influence of oxygen, pressure, heat, pH, heavy metals and other influences that may cause or accelerate degradation [247]. In the case of vitamins, the protection is essential to maintain vitamin levels, while flavor encapsulation is important to avoid unwanted off-taste. Microencapsulation of pigments such as β -carotene is necessary to achieve special physical effects such as high colour strength and special colour hue [247].

As is the case with vitamins, flavours, and pigments, sensitive oils such as fish oil have become increasingly important in the food industry, particularly because the nutritional values. The nutritional benefits of fish oil are generally attributed to their long-chain omega-3 polyunsaturated fatty acids (PUFAs) [137], including both Docosahexaenoic Acid (22:6n-3) or DHA and Eicosapentaenoic Acid (20:5n-3) or EPA [129].

PUFAs have been claimed to have a broad range of beneficial effects including lowering cholesterol, decreasing the risk of arrhythmia, lowering the blood pressure, preventing diabetes in pregnancy, and beneficial effects on joints (relief of arthritis) [185]. Both omega-3 and omega-6 PUFA are precursors of hormone-like compounds, which are involved in many important biological processes in human body [287]. In addition, DHA and EPA play an important role in early infant nutrition particularly for the development of vital human organs such as the neural tube [137]. The imbalance of these PUFAs is believed to cause a variety of diseases [143].

In functional food development, incorporation of PUFAs into food products is dominated by omega-3 fatty acids (α -linolenic acid (ALA) C18:3n-3, eicosapentaenoic acid (EPA) C20:5n-3, docosahexaenoic acid (DHA) C22:6n-3) and omega-6 fatty acids (γ -linolenic acid (GLA) C18:3n-6 and arachidonic acid (AA) C20:4n-6) [6]. Soybean, canola, flaxseed, hemp, and perilla oils are the major sources of ALA, while GLA is mostly found in evening primrose, blackcurrant and borage oils [287]. Oils from the marine algae *Cryptecondium conchii* are mainly rich in DHA only, while fish oil contains both EPA & DHA [287].

Although the nutritional values of fish oil are recognized, adequate daily intake is difficult to achieve. Fish consumption is relatively low in many countries, especially consumption of oily fish with high levels of omega-3 PUFAs [137]. Because of their sensitivity to oxidation, fish oils need to be stabilized to protect them from oxidation. In food application, fish oil interaction with other food ingredients needs to be prevented [6]. Attempts to prevent fish oil oxidation to allow omega-3 fatty acids to fulfill their functions are not trouble-free.

Fish oils in their natural state have a taste and smell that make them less attractive to consumers [185]. Processing technology for masking the smell and taste in food systems faces great challenges. Therefore, to address the problems concerning the susceptibility of fish oil to oxidation and its unpleasant smell, microencapsulation, where the oil is packaged within carrier materials, may be used in place of bulk oils.

Intensive research in the field of fish oil microencapsulation has been done in food industries, food research centres as well as in university laboratories (Table 1.1). A number of coating materials including sodium and calcium caseinate [32;129;142], soy protein [32], whey protein [32;129], gelatine [127], maltodextrin with a wide range of DE [104;129;147;290], sucrose and lactose [127], starches, modified starches [55;274], gum acacia [127], modified cellulose (MC and HPMC) [147], as well as highly branched cyclodextrin (HBCD) [129] have been reported to protect fish oils against oxidation.

Despite types of coating material, Table 1.1 also informed the importance of encapsulation process to products' stability. Thies (2004) classified the encapsulation process as chemical (A) or mechanical (B) processes. A chemical process may rely only on the physical phenomena, while in a mechanical process a chemical reaction may actually be involved. Some typical processes used for producing microcapsules for food application are: (A) complex coacervation, polymer-polymer incompatibility and submerged nozzle processes, and (B) spray drying, spray chilling, fluidised bed coaters, liquid extraction, melt extrusion, suspended nozzles, and spinning or rotating discs [282]. Similarly, Shahidi and Han (1993) divided the technology for forming microcapsules into three groups: physical methods, chemical methods and physicochemical methods [255]. The selection of a method is dependent on economic benefits, sensitivity of core, size of microcapsules, physicochemical properties of both core and wall, application for food ingredients, and release mechanisms.

Literature review indicates that even the best combination of biopolymers for microencapsulating fish oil used with different drying techniques can produce both stable and unstable products. It is necessary to determine which combinations are the best for microencapsulating the right amount of PUFAs in fish oil. To date, there are data gaps in the field of fish oil microencapsulation that need to be filled. No data was found on microencapsulation of fish oil by spray granulation (SG) and application of secondary coating after SG by the fluid bed film coating process (SG-FC). Most importantly, there are no studies where various production methods are compared. The present study therefore is conducted to evaluate and compare fish oil microencapsulation by spray granulation (SG), spray granulation followed by fluid bed film coating (SG-FC), spray drying (SD), and freeze drying (FD). Previous research usually investigated

microencapsulation by a commonly used method such as spray drying or only by freeze drying. Unsatisfactory outcomes obtained by a certain method might be different if produced by other processes. Each drying method offers advantages and disadvantages and so does each coating material.

Selection of the best coating materials and microencapsulation process are crucial steps in food microencapsulation. By carefully examining these two factors, this research is designed to answer the questions associated with the stability of fish oil microcapsules against oxidation in relation to how they are produced.