Fatty Acid Composition and Stability of Extracted Mackerel Muscle Oil and Oil-Polyethylene Glycol Particles Formed by Gas Saturated Solution Process

A. S. M. Tanbirul Haque¹, A.K.M. Asaduzzaman¹ and Byung-Soo Chun*¹

Department of Food Science and Technology, Pukyong National University, 45 Yongso-ro, Nam-Gu, Busan 608-737, Republic of Korea

Abstract

The oil in mackerel muscle was extracted using an environment friendly solvent, supercritical carbon dioxide (SC-CO₂) at a semi-batch flow extraction process and an n-hexane. The SC-CO₂ was maintained at a temperature of 45°C under pressures ranging from 15 to 25 MPa. The flow rate of CO₂ (27 g/min) was constant during the entire 2 h extraction period. The fatty acid composition of the oil was analyzed using gas chromatography (GC). Significant concentrations of eicosapentaenoic acid (EPA) acid and docosahexaenoic acid (DHA) acid were present in the SC-CO₂ extracted oil. The oil extracted using SC-CO₂ exhibited increased stability compared with n-hexane extracted oil. Particles of mackerel oil together with the biodegradable polymer, polyethylene glycol (PEG) were formed using a gas saturated solution process (PGSS) with SC-CO₂ in a thermostatted stirred vessel. Different temperatures (45-55°C), pressures (15-25 MPa) and a nozzle size 400 µm were used for PGSS with a 1 h reaction time. The stability of mackerel oil in the particles did not changed significantly.

Keywords: Docosahexaenoic acid, Eicosapentaenoic acid, Mackerel oil, Oil stability, Polyethylene glycol, Supercritical carbon dioxide.

Introduction

Fish oil is derived from the tissues of oily fish, it is recommended as part of healthy diet because it contains ω-3 polyunsaturated fatty acids (PUFAs), such as eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA). The beneficial health effects of ω-3 PUFAs are well defined: they are essential for the normal growth and development of the brain and nervous system and are also thought to exert beneficial effects during the treatment of coronary artery disease, hypertension, arthritis, clinical depression, anxiety, inflammatory and autoimmune disorders and cancer (Cao and Hur, 2005; Correa et al., 2008; Jeong et al., 2006; Lee et al., 2006; Su et al., 2003; Naliwaiko et al., 2004; Green et al. 2006; Yehuda et al., 2005; Nemets et al., 2002).

Mackerel belongs to the family Scombridae, and is abundant in cold and temperate shelf areas. It is a fatty species and the fat content is well-distributed throughout the body (Osman et al., 2001). Mackerel also contains significant amounts of protein, essential amino acid, lipid and many other biologically active compounds.

Several methods have been reported for extracting fish oils that result in varying yields. Lipids are conventionally extracted and purified using methods such as hexane extraction, vacuum distillation, urea complexation, or conventional crystallization. However, these methods have the disadvantage of requiring high-temperature processing those results in the decomposition or degradation of thermally labile compounds or of employing toxic solvents with adverse health effects (Hultin, 1994; Staby and Mollerup, 1993). Supercritical carbon...
dioxide (SC-CO₂) extraction is a novel and promising process for the extraction and fractionation of edible oils containing labile PUFA's. Using carbon dioxide as the solvent is advantageous, because it is non-flammable, non-toxic, inexpensive, and can be used under mild operating conditions.

The formulation of natural substances together with a biocompatible or biodegradable carrier material to form composites or encapsulates has great potential for the pharmaceutical, cosmetic and food industries (Cocero et al., 2009). Natural substances such as carotenoids, fatty acids and antioxidants are being used extensively in a variety of food products (Budavari, 1989). In addition several clinically approved pharmaceutical products use biodegradable polymers to regulate the rate of drug release within the body (Tracy, 1998; Okada, 1997).

Different processes have been used for encapsulation, including spray-drying, freeze-drying, liquid antisolvent crystallization and milling processes. However, there are several disadvantages to these technologies, such as the production of coarse particles with a broad particle size distribution, product degradation due to mechanical or thermal stress and particle contamination with organic solvents or other toxic substances. Therefore, novel alternative precipitation methods are currently being investigated (Martin and Cocero, 2008).

Particle formations techniques SC-CO₂ such as the rapid expansion of supercritical solutions (RESS), particles from gas saturated solutions (PGSS), and supercritical anti-solvent (SAS) precipitation have received much attention as precipitation methods alternative to those using organic solvents (Mishima, 2008). These methods are important for drug delivery systems to successfully obtain composites or encapsulates that comprise an active compound loaded into a matrix of a carrier material, thus improving product preservation as well as controlling the dissolution rate of the active compound (Cocero et al., 2009).

Achieving small particles with a narrow particle size distribution for pharmaceutical agents is a major aim in the design of conventional drug delivery systems such as tablets, capsules, injection. Biphasic drug delivery systems such as suspension and emulsion and controlled drug delivery systems such as implants, transdermal, microemulsions and nanoparticles are also important for pharmaceutical development (Budavari, 1989; Mishima, 2008; Turk and Lietzow, 2008; Yildiz et al. 2007; Park and Yeo, 2008; Tandya et al. 2006; Li et al., 2006).

PGSS can be used to produce microparticles with a narrow size distribution; therefore, it is a key technique used in the food and pharmaceutical industries, because it results in solvent-free products (Pathak et al., 2006). Therefore, the aim of this study was to extract mackerel oil using SC-CO₂ and hexane to compare the fatty acid composition and stability of extracted oil and oil particle.

Materials and Methods

Materials

Mackerels were collected from the Busan Cooperative Fish Market (Seo-gu, Busan, Korea). The muscle was separated mechanically and then washed thoroughly with cold distilled water in the laboratory. Pure carbon dioxide (99.99%) was supplied by KOSEM (Sangbuk-myeon Yangsan, Korea). All other chemicals used in this study were of analytical or HPLC grade.

Sample preparation

The mackerel muscle was dried in a freeze-dryer for about 72 h and then crushed using a mechanical blender supplied by DONG YANG PCS CO. LTD. (Ansan, Korea). These “freeze dried mackerel muscle” samples were then stored at -20°C.

SC-CO₂ extraction

Laboratory scale of supercritical fluid extraction (SFE) was performed. 20 g of freeze dried raw mackerel muscle was loaded into a 200 mL stainless steel extraction vessel. A thin layer of cotton was placed at the bottom of the extraction vessel and a second layer was placed above the sample before plugging the cap. CO₂ was pumped into the extraction vessel at a constant pressure using a high pressure pump (Milroyal, Milton Roy, USA) until the desired pressure was obtained. A backpressure regulator was used to control the CO₂ pressure. The extraction temperature was maintained by connecting the extraction vessel to a water bath. Flow rates and the accumulated gas volume passing through the apparatus were measured using a gas flow meter (Shinagawa, Tokyo, Japan). After SC-CO₂ extraction, the remaining mackerel muscle residues in the vessel and oil was stored at -20°C until further use and analysis. Mackerel muscle was extracted at a temperature of 45°C and pressure ranging from 15 to 25 MPa for 2 h using SC-CO₂. The flow rates of CO₂ were kept constant at 27 g/min for all extraction.

Hexane extraction

Extraction was performed using hexane as the solvent. 40 g of freeze dried raw mackerel muscle were placed into a beaker with 200 mL hexane and stirred at 300 rpm for 20 h at 45°C. After extraction, the hexane solution was filtered using a filter paper and then evaporated in a rotary vacuum evaporator at 40°C. The oil was then stored at -20°C until use.

Particle formation using PGSS

The experiments were carried out using PEG 8000 (g/mol) and mackerel oil at different pressures and temperatures. A