Int.J.Curr.Microbiol.App.Sci (2020) *9*(12): 730-735

International Journal of Current Microbiology and Applied Sciences
ISSN: 2319-7706 **Volume 9 Number 12 (2020)**
Journal homepage: http://www.ijcmas.com

Review Article https://doi.org/10.20546/ijcmas.2020.912.088

Supercritical Fluid Extraction of Fish Oil – Recent Perspectives

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Keywords

Supercritical fluids, Extraction efficiency, DHA, Fish oil

Article Info

Accepted:
07 November 2020
Available Online:
10 December 2020

A B S T R A C T

Health benefits of omega-3 polyunsaturated fatty acids, such as Eicosapentaenoic acid (EPA) and Docosahexaenoic acid (DHA) has been well documented by researchers worldwide. Fish oil is reported to be one of the richest dietary sources of EPA and DHA. As a result, fish oil is being extracted on a wide scale globally using several methods. Recently, several innovative and sustainable techniques have been put forth by researchers for extraction of good quality fish oil. Supercritical fluid extraction is being recognised as one of the most feasible method for extracting high quality fish oil. The technique has been used extensively for extracting oil from fish meat as well as processing discards. The present review details on the extraction of fish oil using supercritical fluid extraction.

Introduction

Fish oil is often considered as one of the richest and relatively inexpensive source of omega-3 polyunsaturated fatty acids, especially EPA and DHA. EPA and DHA are known for their bioactivities such as prevention of cardiovascular diseases, inflammation, tumors and for the functioning of brain, liver, heart (Agostoni, 2008; García-Almeida *et al.,* 2010). The role of DHA in visual development of foetuses and infants has been also reported (Arab-Tehrany *et al.,* 2012). Apart from the major omega-3 fatty acids, fish oil is also considered to be rich in fat soluble vitamins, such as vitamin A and D. Several health organisations has recommended consumption of fish and fish oil for overall wellbeing of humans. World Health Organization recommends about one to two serving of fish consumption weekly which corresponds to 200–500 mg of EPA+DHA (Kris-Etherton *et al.,* 2009). Furthermore, the international Society for the Study of Fatty Acids and Lipids suggests daily intake of about 500 mg EPA and DHA can help in prevention from cardiovascular diseases (Cunnane *et al.,* 2004). The increase in consumer awareness regarding the nutritional richness of fish oil has further fostered its extraction for human consumption. Several extraction methods are being employed for production of high quality fish oil. Wet reduction, enzymatic assisted extraction, etc. are some of the commonly employed methods for obtaining fish oil. Among the conventional methods wet reduction is the most commonly employed method, which involves three processing steps: cooking at temperature followed by pressing and centrifuging. However, such methods have serious setbacks in terms of the quality of oil obtained as high temperature employed for the extraction process result in degradation or decomposition of the thermally labile compounds resulting in production of oxidised oils. Recently, several sustainable extraction methods have been developed for extraction of fish oil such as supercritical fluid extraction (SFE), microwave assisted extraction, ultrasound assisted extraction, enzymatic assisted extraction etc.

Supercritical Fluid Extraction (SFE)

Supercritical fluid extraction is similar to any other extraction technique; the only difference here is the use of supercritical fluids as solvents for extraction purposes. Supercritical fluids (SCF) are those solvents which possess both liquid and gas-like properties when the temperature and pressure are above their critical point (Khaw *et al.,* 2017). The low viscosity combined with high diffusion enables the rapid diffusion of solvent into the matrix and thereby facilitating in the extraction process. Among the different SCFs available for extraction, supercritical carbon dioxide (SC-CO_2) is the most commonly employed solvent (Duarte *et al.,* 2014). The easily attainable critical conditions (critical temperature: 31.1°C, critical pressure:72.8 bar) along with its non-toxicity and easy availability has made SC-CO_2 as the most desirable solvent for SFE (Espinosa-Pardo *et al.,* 2017; Wrona *et al.,* 2019). The highly non-polar nature of SC-CO_2 makes it most favourable for extraction of non-polar compound. The use of polar solvents along with SC-CO_2 will change the polarity of the latter, and thereby makes it useful for extraction of polar compounds too (Garcia-Mendoza *et al.,* 2017). The polar solvents which are used to change the polarity of SCFs are often referred to as co-solvents or modifiers. The most commonly employed co-solvent is ethanol as it is food-grade and easily available.

SFE for extraction of fish oil

The production of high quality fish oil has assumed great significance in the recent past owing to its reported health benefits. Production of good quality fish oil depends to a greater extent on the type of raw material, quality of raw material, the type of extraction method and extraction conditions. Globally, there is an increasing trend to utilise the fish/seafood processing discards as raw material for production of high quality bioactive. It has been well documented that both fish and fish by-products can serve as excellent resources for extraction of omega-3 rich oil. SFE has evolved as one of the most feasible sustainable extraction methods for obtaining fish oil (Létisse *et al.,* 2006). Rubio-Rodríguez *et al.,* (2012) have compared four different methods, such as cold extraction, wet reduction, enzymatic extraction and supercritical fluid extraction for obtaining oil from fish by-products. It was observed that among all methods, SFE can be a feasible method from the stability and safety point of view, as fish oil extracted by SFE had better oxidative stability and reduced arsenic content. Sahena *et al.,* (2010) has compared soxhlet extraction with SFE for obtaining oil from mackerel skin. It was observed that almost similar yield of fish oil was obtained using both the methods. Similarly, Sahena *et al.,* (2010) has analysed the fatty acid profile of oil extracted from various parts of Indian mackerel (*Rastrelligerkanagurta*) such as skin, flesh, viscera and head using various techniques of supercritical carbon dioxide

($SC-CO_2$), viz, continuous, cosolvent, soaking and pressure swing. All the extractions were carried out the same conditions (350 bar pressure, 60 °C temperature) and the efficiency of the process was compared with that of soxhlet. It was reported that among the different techniques employed, soaking and pressure swing techniques were efficient in extracting major polyunsaturated fatty acids, EPA and DHA.

Generally, the extraction efficiency of SFE are reported to be affected by change of major factors such as pressure, temperature, time, co-solvents etc. (Plaza and Rodríguez-Meizoso, 2013). Several researchers have investigated the effect of these variables on the yield, quality and quantity of bioactives obtained. Létisse *et al.*, (2006) have optimised the extraction conditions for obtaining EPA and DHA rich oil from sardine and efficiency was compared with that of conventional solvent based extraction. It was reported that about 11% of EPA and 13% of DHA were extracted from sardine at the optimised extraction conditions (300 bar pressure, 75 °C temperature, 45 min time). Though the extraction yields were better with solvent based extraction, it was suggested that SFE can be more advantageous from a time and quality point of view.

Sarker *et al.*, (2012) have investigated the potential of SFE in valorisation of viscera of African Catfish and compared with that of soxhlet method. A yield of about 67% was recorded at the optimized extraction condition of 400 bar pressure, temperature 57.5 °C for an extraction period of 150 min which was reasonable with that of the yield obtained by soxhlet method. It was also reported that the major operating variables such as pressure, time, temperature and flow rate was found significant in the extraction process.

Table.1 SFE for extracting fish oil

Sl No:	Samples used for extraction	References
1.	**Fish by products**	
a.	Salmon Roe	Tanaka*et al* (2004)
b.	Sardine Heads	Létisse*et al*., 2006)
c.	Skin of Indian Mackerel	Sahena*et al* (2010)
d.	Off cuts from fishes, squid liver	Rubio-Rodríguez *et al* (2012)
e.	Viscera of African Catfish	Sarker*et al* (2012)
f.	Skin, muscle tissue, and Viscera of Mackerel	Hajeb*et al* (2015)
g.	Fillets, viscera and caviar of Common Carp	Kuvendziev*et al* (2018)
2.	**Tuna by products utilization**	
a.	Tuna red meat	Yathavamoorthi*et al* (2015
b.	Head, skin and viscera	Ferdosh*et al* (2015)
c.	Skin, scale and bones	Ahmed*et al* (2017)
d.	Tuna Liver	Fang *et al* (2019)

The potential of SFE in utilising tuna meat and its by-products has also been reported. Yathavamoorthi *et al.*, (2015) have employed SFE for extraction of oil from freeze dried tuna red meat, which is a major by-product from tuna processing industry. It was reported that about 5% oil was obtained using the process, which was rich in polyunsaturated fatty acids like EPA and DHA. They have suggested that SFE can be effective process in utilizing tuna red meat without incurring much yield losses. Hsieh *et al.,* (2005) have utilised SFE for concentrating EPA and DHA ethyl esters from tuna cooking juice. They have reported that factors such as temperature and pressure had influenced the extraction efficiency, that is to separate EPA and DHA esters. Similarly, Fang *et al.,* (2019) have attempted to utilize tuna liver using various methods such as subcritical dimethyl ether extraction, wet reduction, enzymatic extraction and supercritical fluid extraction. Among all techniques employed, both subcritical dimethyl ether extraction and SFE were found to be the most effective as these techniques can prevent the oxidation of oil and vitamins extracted and thereby yielding a high quality oil. Ahmed *et al.,* (2017) have studied the quality of oil obtained from different parts (skin, scales, bones) of big eye tuna (*Thunnusobesus*) using SFE and compared with that of the oil extracted by conventional hexane based extraction. Significant findings of the study suggested that oil extracted by SFE can be considered as high quality as revealed by various physico-chemical attributes. Color and viscosity were better in SFE extracted oil. Similarly, other oxidative stability parameters such as anisidine value, peroxide value, free fatty acids, paraanisidine, TOTOX values has also shown oil extracted using SFE better oxidative stability. In addition, the heavy metal content which got extracted during the extraction process were less in SFE extracted oil. It was hence concluded that SFE can be a better process to produce oxidative stable and safe fish oil. Ferdosh *et al.,* (2015) have also reported similar results stating SFE can be a feasible produce good quality oil from tuna processing discards when compared to that of the conventional methods. Table 1 below shows the sources used for oil extraction recently.

In conclusion the fish, a rich repository of biomolecules is often explored for production of omega03 fatty acids. Several techniques are being employed for its production. Recently, greener extraction methods are being for recovery of biomolecules from fish and seafood processing discards. Supercritical fluid extraction technique is being widely explored for biomolecules recovery from wide sources presently. The technique has many advantages when compared to the conventional methods, most importantly, the ambient operating conditions which can ensure production of good quality fish oils. However, the operating costs for SFE are high at a production scale. Apart from this, the raw material used for SFE extraction should have less moisture content (below 20%), demanding that is should go pre-treatments before the actual process. Hence, newer innovations has to be introduced to make the technique more economically and commercially feasible too.

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How to cite this article:

Anas, K. K., Lekshmi R. G. Kumar, C. S. Tejpal and Suseela Mathew. 2020. Supercritical Fluid Extraction of Fish Oil – Recent Perspectives. *Int.J.Curr.Microbiol.App.Sci.* 9(12): 730-735. doi: https://doi.org/10.20546/ijcmas.2020.912.088