学位論文内容の要旨

博士の専攻分野名称：博士（農学）　　氏　名：Disanayakage Surakshi Wimangika Rajapaksha

学位論文題名

Integrated approach for valorisation of polyphenols in spent black tea: extraction, microencapsulation, and development of functional packaging film

（紅茶殻ポリフェノール類高付加価値化のための統合的アプローチ：抽出，マイクロカプセル化，機能性包装フィルムの開発）

In a circular economy, the utilization of food manufacturing waste for obtaining functional ingredients has received great attention. Spent black tea (SBT) is an abundant waste generated after beverage manufacturing process and considered as a potential and underutilized source of polyphenols with high antioxidant power. Moreover, the studies which utilized the SBT for the recovery of polyphenols and their subsequent use as a natural antioxidant in the food packaging have not been reported to date. Hence, the main objectives of the research are: (1) to develop a method for recovering phenolic compounds from SBT by integrating the processes of subcritical solvent extraction (SSE) and microencapsulation using different wall materials by spray drying technique (2) to assess the potential of recovering polyphenols in SBT by SSE at pilot-plant scale to propose a suitable scale-up method, and (3) to develop and characterization of a functional food film incorporating SBT extract.

1. Valorisation of spent black tea by recovery of polyphenols: subcritical solvent extraction and microencapsulation

The first study evaluated the SSE for recovery of polyphenols in SBT, and microencapsulation to improve the stability of obtained extract. Optimization of extraction conditions was carried out by response surface methodology for the best recovery of antioxidant phenolic compounds. Two variables [temperature (°C) and ethanol concentration (%)] were used to design the optimization model using central composite inscribed. Extraction temperature of 180°C and ethanol concentration of 71% were optimal for the highest yield of total polyphenols (126.89 mg gallic acid equivalent/g SBT) and antioxidant activity (69.08 mg gallic acid equivalent/g SBT). The extract was encapsulated using pectin, sodium caseinate, and a blend of these compounds (ratio 1:1) as wall materials by spray drying. The blend of wall materials produced an amorphous powder with the highest

phenolic retention (94.28%) in the accelerated storage at 45°C for 40 days. Obtained microencapsulates have potential use in different food systems to enhance their antioxidant property.

2. Pilot-scale extraction of polyphenols from spent black tea by semi-continuous subcritical solvent extraction

By exploiting the lab-scale knowledge, a pilot-scale process on semi-continuous SSE of polyphenols from SBT was developed. Treatment of SBT with ethanol-water (50% w/w) as solvent at 125 °C and 0.3 MPa achieved a significantly higher yield of polyphenols (80.82 g gallic acid equivalents/kg black tea) with antioxidant activity (64.20 g gallic acid equivalents/kg black tea), compared to hot water extraction (HWE). SSE increased the soluble matter content in extracts than HWE. Based on the results of LC-MS, theaflavin-3,3'-digallate was the most abundant polyphenol from a total of 12 compounds to be extracted by SBT with 50% ethanol. The results suggested that SSE can be used as a scale-up extraction method to recover polyphenols from SBT.

3. Development and characterization of functional starch-based films incorporating free or microencapsulated spent black tea extract

The potential of using SBT extract as an active ingredient in food packaging was evaluated. Free or microencapsulated forms of SBT, using a pectin–sodium caseinate mixture as a wall material, were incorporated in a cassava starch matrix and films developed by casting. The effect of incorporating SBT at different polyphenol contents (0.17% and 0.34%) on the structural, physical, and antioxidant properties of the films, the migration of active compounds into different food simulants and their performance at preventing lipid oxidation were evaluated. The results showed that adding free SBT modified the film and created a less elastic film with antioxidant activity. Incorporating microencapsulated SBT improved the mechanical properties of active films and preserved their antioxidant activity. Both types of active film delayed lipid oxidation up to 35 d. This study revealed that starch film incorporating microencapsulated SBT can be used as a functional food packaging to protect fatty foods from oxidation.

4. Conclusions

Subcritical solvent extraction was an efficient method for extracting polyphenols from SBT. Microencapsulation turned the SBT extract into valuable food ingredient with improved stability. The SBT extract had the potential to be incorporated into cassava starch in developing packaging films with improved antioxidant activity, which allows the potential application in the prevention of lipid oxidation of fatty foods. Further quantification of phenolic compounds will ease the better identification of all extracts. Moreover, further studies are required to increase the mechanical properties of functional films.