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学位の種類	博士（工学）
学位記番号	甲第１７３号
学位授与年月日	平成１８年　３月１８日
学位授与の要件	学位規則第４条第１項該当
学位論文題目	Influence of Emulsion Size and Powder Morphology on the Microencapsulation and Stability of Spray-Dried Flavors （乳化フレーバーの噴霧乾燥および安定性に与える乳化粒子径および乾燥粒子形状の影響）
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学位論文の内容の要旨

Microencapsulation of flavors is a technology of enclosing flavor compounds in a carrier matrix to provide dry and free-flowing powders. It is also important to provide protection against the degradative reaction and to prevent the loss of flavors during food processing and storage. Spray drying is the most common technique to produce flavor powders. For the hydrophobic flavors, the flavors were emulsified in the carrier solution, which usually composed of gum arabic, hydrolyzed starch, modified starch or soy protein, to form an emulsion. The emulsion is then fed into the spray dryer. First, the mixture of core and wall materials is transformed into droplets by an atomizer. In the second, the hot air flowing contacts the atomized droplets and evaporates the water. The dried particles, consisting of dry matrices in which the core material is held in a micro-dispersion, fall through the gaseous medium to the bottom of the dryer and are collected. In recent 4 decades, numerous studies have been done in these processes. Recently, the main emphasis of the microencapsulation of flavors has concentrated on preventing the flavor losses during spray drying and extending the shelf life of the products.

In this research, the goal is to understand the mechanism loss of flavor during spray drying process in order to improve the flavor retention. The focus was on the effects of emulsion droplet size, powder size as well as the type of the model flavors and the wall materials. More specifically the aim is to understand the mechanism of the loss and oxidation of encapsulated flavor in the particle which directly relates to the shelf life of product.

The work also focused on the effect of water activity as well as the change of the capsule structure on the stability of encapsulated flavor. Furthermore, in order to understand the morphology of spray-dried powder and encapsulated flavor powder, confocal laser scanning microscopy (CLSM) was used to view the cross sectional image of the spray-dried powder and the arrangement of encapsulated flavor inside powder without the destruction of the powders.

d-Limonene, ethyl butyrate, and ethyl propionate were used as the model flavors to study the influence of the mean emulsion droplet size on the flavor retention during spray drying. The increasing emulsion droplet size decreased the retention of flavors. The larger emulsion droplets would be changed in size during atomization, resulting in the decreasing flavor retention. For the water-soluble flavors, ethyl butyrate and ethyl propionate, the retention had a maximum at the optimum value of a mean emulsion diameter. Further, the powder size seemed to give a less important effect on the flavor retention, as compared to the influence of emulsion size.

The stability of encapsulated flavor was studied in view of the release characteristics and oxidation stability. The water activity played an important role on the stability of the flavor. In the case of the water-insoluble flavor, *d*-limonene, the release and oxidation rates have a minimum at the water activity of around 0.70-0.80. That could be explained by the change of capsule structure of the wall materials. The release and oxidation rate constants could be correlated with the difference between the storage temperature and glass temperature. On the other hand, the partially soluble flavor (*i.e. l*-menthol), the release rate increased monotonously with the increasing water activity. This might be caused by the solubility of the *l*-menthol. For partially water soluble flavors, dissolved flavors can increasingly diffuse through the wall matrices with increasing of water activity. The release rate was controlled by the water adsorption of wall matrices and the solubility of flavor.

The stability was also investigated with respect to the emulsion droplet size and powder particle size. The release and the oxidation decreased deeply with an increase in powder and emulsion droplet size. Further, the distributions of emulsion size during storage showed an increase in the fraction of large emulsion droplets and changed to a bimodal distribution. However, the modified starch showed a higher stability of encapsulated *d*-limonene than the others. The influence of powder and emulsion size on its encapsulation efficiency as well as the change in the emulsion size during storage could not be observed.

Finally, confocal laser scanning microscopy showed new challenges for studying the morphology of spray-dried powder and encapsulated flavor powder without the destruction of their structures.

論文審査の結果の要旨

噴霧乾燥法は液状原料を微小液滴に噴霧し、これを高温空気で瞬時に乾燥する手法であり、機能性食品の製造法として多用されている。しかしながら、乾燥中の有用成分の熱劣化や、保存中の安定性に関する研究は極めて定性的なものが多いのが現状である。本研究は、機能性フレーバー粉末作製における、噴霧乾燥中のフレーバー散失に与える諸因子の影響と、長期保存中のフレーバー徐放機構の解明を目的としたものである。

食品フレーバーは難水溶性のものが多く、これを乾燥粉末とするために、乳化剤などを用いてミクロンオーダーの微小油滴を含む液（エマルション）を作製し、これを噴霧乾燥した。乾燥中のフレーバー残留率は、エマルションの大きさに顕著に依存することを見いだした。この原因として、回転円盤式噴霧器の高剪断場におけるエマルションの剪断破壊と、フレーバーの僅かな水溶性が原因であることを明らかにした。次いで、粉末フレーバーの保存中における徐放と酸化実験を種々の湿度環境で行い、徐放速度および酸化速度を乾燥粒子とエマルションサイズ、および糖質マトリックスの構造因子と関連づけて解析した。徐放や酸化などのフレーバー保存安定性は、乾燥粒子サイズによって顕著に影響されること及び、徐放速度と酸化速度が共に糖質マトリックスのガラス転移温度近傍で極大値を示すことを明らかにした。噴霧乾燥粒子は内部に空洞を有する中空粒子となることが従来から指摘されている。本研究では、共焦点レーザー顕微鏡を用いて乾燥粒子の断面蛍光像を得ることに成功し、乾燥条件による粒子形状変化を非破壊状態で研究できる可能性を示した。更に、油溶性蛍光剤をフレーバーに添加してエマルションを作製することにより、噴霧乾燥粒子中のエマルションの可視化にも成功した。これらの新技術は機能性フレーバー粉末の機能特性評価を行う上で極めて有用であると考えられる。

以上、本論文の研究成果は、噴霧乾燥による機能性食品粉末の開発・工業化および評価を行う上で極めて重要であり、この分野の研究の発展に大いに貢献できるものと考えられる。

よって、本論文は博士（工学）の学位論文に値するものと認められる。