The Emulsifying Performance of Mildly Derived Extracts from Argan By-products: Towards a Sustainable Production of Natural Emulsifiers （アルガン副産物からの抽出物の乳化性の解析と応用　-天然乳化剤の持続的生産をめざして-）

著者（英）	Noamane Taarji
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氏　　名	Taarji Noamane
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	（職名）	（学位）	（氏名）
主査	筑波大学准教授	博士（農学）	Marcos Antonio das Neves
副査	筑波大学教授	博士（農学）	礒田　博子
副査	筑波大学特命教授	工学博士	中嶋　光敏
副査	筑波大学教授（協働大学院）	博士（農学）	小林　功

Abstract of thesis

Argan oil extraction industry generates every year large amounts of by-products that are not efficiently valorized. On average, 1 ha of argan trees can produce around 300 kg of dried fruits, in the form of 20 kg of kernels and only 8 kg of oil. This does not only result in the loss of potentially high value-added compounds from these materials but also impacts the local environment, through overexploitation of argan trees for fruit harvesting and oil extraction. Regarding this, a systematic study of the secondary metabolites of argan tree was initiated since the early 90's. The objective of this study was to identify new bioactive compounds that can increase the economic and industrial values of argan trees. The results of this study allowed to characterize, within the different parts of argan tree, a wide variety of functional compounds, with some of them already evaluated for different biological activities.

In the introductory section, the author presented an overview of the different valorization opportunities of argan by-products in cosmetic and pharmaceutical applications. The great potential of obtaining natural emulsifiers from agro-industrial by-products was also pointed out. The aim was to suggest the use of argan by-products as a source of natural emulsifiers. The rich surface-active composition of these materials (e.g.

proteins, saponins), as well as their significant generation quantities, can create potential applications as natural emulsifiers.

Next, the surface-active and emulsifying properties of saponin-rich extracts from argan oil press-cake was evaluated. The aim was to produce model oil-in-water (O/W) nanoemulsions using these extracts as sole emulsifiers. Various extracts were initially prepared in order to select the most surface-active ones foreseeing emulsions preparation. Fifty percent (v/v) ethanolic extract reduced the interfacial tension to a minimum value at both medium chain triglycerides (MCT) oil or soybean oil and water interfaces. This extract was also effective at producing stable nanoemulsions using different oils such as soybean oil, MCT oil or fish oil, and with similar properties to those obtained by conventional emulsifiers such as Tween 20. The physical stability and biological fate of the emulsions prepared using argan extract were investigated. The emulsions were very sensitive to salt addition more than 25 mM and to extreme acidic pH lower than 3, indicating that the main stabilization mechanism is electrostatic, likely due to the presence of surface-active compounds with ionizable groups, such as saponins. The emulsions were also very sensitive to gastric conditions, particularly when pepsin was added to the digestion system, which highlights the contribution of proteins to the surface-active and emulsifying properties of 50% ethanolic extract.

Then, the emulsifying performance of argan extract was evaluated using microchannel emulsification (MCE). The aim was to produce stable monodisperse O/W emulsions using the extract a sole emulsifier. The complex composition of this extract imparted its emulsifying efficiency, by creating a hydrophobic, or slightly hydrophilic, layer on the MC array plate surface. This resulted in unsuccessful emulsification using short MCs of 70 μm, but did not affect the emulsification efficiency in longer ones. Using these longer channels with 160 μm, the author could produce stable monodisperse O/W emulsions, with similar droplet size and droplet size distribution to those stabilized by Tween 80, and for up to 10 h of continuous emulsification. The author also compared the surface-activity and emulsifying properties of a crude extract from licorice root and purified saponins from the same origin. The aim was to examine the contribution of specific compounds to the emulsifying performance of multicomponent plant extracts. As expected, the non-purified extract was more surface-active and effective at producing small droplet size emulsions, in comparison to purified saponins. The emulsions priduced were superiorly stable at low pH and high salt concentrations, but less stable at elevated temperatures, suggesting again the contribution of proteins to the observed results. Evidences presented in this chapter indicated that non-purified surface-active extracts, such as argan and licorice root extracts, can provide superior emulsifying ability in comparison to purified emulsifiers. This is due to the contribution of multiple compounds to the overall surface-active and emulsifying properties, and most likely the formation of biogenic complexes between these compounds at the oil/water interface, thus leading to improved droplet coverage and stability.

Finally, the author suggested innovative scenarios for the valorization of argan by-products. Sequential

extraction methods should be adopted in the future to simultaneously obtain various bioactive and dietary compounds from these materials. Suitable storage conditions, innovative harvesting methods and adjusted oil production chains should be also considered in order to preserve specific natural compounds in the different parts of argan fruit, without affecting the productivity of the tree. For successful application in the food and beverage industries, other parameters such as bitterness, potential toxicity, cost and supply reliabilityare important.

Abstract of assessment result

【Review】

Emulsifiers are one of the most utilized ingredients in food industry. They are ubiquitous in many food products, in which they provide multiple properties, such as good dispersibility, prolonged stability and/or improved bioavailability of other ingredients. Many natural emulsifiers are currently available in the market. Among these, proteins, lecithin and polysaccharides are relevant. The increasing demand of consumers towards 'green label' products have attracted however additional interest in identifying other sources of natural emulsifiers. Particularly, the use of crude plant emulsifying extracts, obtained via simple extraction/separation steps, can be of great interest. In this study, the author evaluated the interfacial and emulsifying properties of various aqueous-ethanolic extracts from argan by-products. The purpose of the study was to produce stable emulsions using these extracts as sole emulsifiers. To provide clear insights about their emulsifying mechanism in O/W emulsions was expected.

The results showed that it is possible to produce stable O/W emulsions using argan extracts as sole emulsifiers. The stabilization mechanism is believed to be exclusively electrostatic, likely due to the formation of biogenic complexes between proteins and saponins. The emulsions had similar physicochemical characteristics to those prepared using conventional emulsifiers and were stable up to two months of storage at different temperatures. Nevertheless, for successful applications in the future, it would be important to extend this work by evaluating other properties such as the taste profile and the potential toxicity of the prepared extracts.

【Result】

The final examination committee conducted a meeting as a final examination on June 7, 2019. The applicant provided an overview of dissertation, addressed questions and comments raised during Q&A session. All of the committee members reached a final decision that the applicant has passed the final examination.

【Conclusion】

Therefore, the final examination committee approved that the applicant is qualified to be awarded Doctor of Philosophy in Food Innovation.