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## 学位論文の概要及び要旨

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目<u>Crystal transformation from hydrous to anhydrous crystals with dehydration in ethanol</u> (エタノールを用いた脱水による含水結晶から無水結晶への結晶変換)

学位論文の概要及び要旨

Sugars are among the most important major components in food. The following investigation covers "phase transition of food" from hydrous crystal sugar to anhydrous crystal sugar. This study includes: (1) the concept and molecular arrangement of hydrous crystal sugar or amorphous solid to the formation of anhydrous crystal sugar, and (2) the concept of physical property changes and molecular mobility around the glass transition and mathematical descriptions of crystal transformation and influences on food stability.

There will be a focus on the crystal transformation of sugars related to amorphous solids. Diffusion of water in foods is important in product development, processing and storage design. The rate of crystal structure changes often depends on the physical state and therefore on temperature and water content.

Trehalose is a non-reducing disaccharide found in many organisms where its primary roles are as an energy source and protection of living cells, proteins, and phospholipids from the damage caused by dryness by changing into various crystalline forms. In Tottori University, the crystal transformation from hydrous trehalose to anhydrous trehalose was investigated by the dehydration in ethanol as five month internship research training in ENSBANA. By this new dehydration method, porous crystal structure could be obtained. Consequently to this research, the main object of my study is "Crystal transformation from hydrous to anhydrous crystals with dehydration in ethanol".

The sub topics of my research are :

(1) Kinetic analysis of the crystal transformation from hydrous crystal sugar to anhydrous crystal sugar by the solvent dehydration by Differential Scanning Calorimetry (DSC).

(2) Characterization of anhydrous  $\beta$ -maltose.

(3) Dehydration of amorphous sucrose by ethanol.

(4) Formation of stable anhydrous porous monosodium glutamate crystals, these crystals having the ability to form a creamy gel.

(5) Formation of non-oil creamy gel of sugar using the newly obtained porous crystals.

The time courses of dehydration and crystal transformation were markedly dependent on the operation conditions such as the temperature of ethanol, its initial water content, and stirring rate. The crystal structure of anhydrous sugar crystal such as trehalose is also investigated by Scanning Electron Microscopy (SEM). The anhydrous trehalose by the ethanol dehydration presented a porous structure, implying that it could be applied as an encapsulation matrix for food oils and enzyme. The kinetic analysis from hydrous crystal sugar to anhydrous crystal sugar by Differential Scanning Calorimetry (DSC) is the first step of the study. The hydrous crystals of sugars are placed in a DSC pan with ethanol, and DSC is used as a process (Time, Temperature). The resulting sugar crystals are subjected to several analyses. In case of maltose, the equilibrium between  $\alpha$ -anomer and  $\beta$ -anomer is very important to determine the type of crystal. We cannot obtain  $\beta$ -anhydrous maltose by usual method. The ethanol method will be applied to maltose, and investigate the possibility of obtaining  $\beta$ -anhydrous maltose. The characterization of this new  $\beta$ -anhydrous maltose is important. The surface structures of sugar crystals are analyzed using SEM. The porous structure can be observed. Crystal polymorphism is analyzed by using DSC. Several endothermic peaks are obtained, and analyzed thermo-physically. Powder x-ray diffractogram analysis is used to determine the crystal structure of the sugars, and identifies the category of the crystal polymorphism. Confocal Laser Scanning Micrometer (CLSM) might be key technique to evaluate the porous structure inside the sugar crystal. The pore size can be measured by a Mercury Porosimeter, which can characterize the pore size distribution of the porous sugar obtained by ethanol method. The study includes determination of parameters influencing on the crystal transformation. The transformation rate is analyzed with Avrami's equation, which can often be used for the chemical reaction analysis. The crystal transformation path from hydrous to anhydrous state by the ethanol method might also occur by way of the amorphous state. The dehydration of amorphous sucrose with ethanol is one of the objectives of the study. Crystal transformation via ethanol dehydration will be applied to monosodium glutamate (MSG), a food additive responsible for the umami taste, in order to obtain a stable and porous anhydrous crystal of MSG. Finally, after having obtained porous anhydrous crystals of carbohydrates and MSG, the formation of creamy gel from the newly obtained crystals will be investigated by rehydration. The formation of a non-oil gel of sugar or MSG would be of great interest for the food industry.