

# Expansion of the cavity in the sago starch crystalline structure by water molecules

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**Abstract:** X-ray diffraction, one of the useful tools for the structural analysis of starch, was used for analysis of the reflection surface of a crystalline structure, resulting in determining the distance of the interlayered structure. The X-ray diffraction peak at about 5 to 6° for  $2\theta$  Cu  $K\alpha$  of water-added starch with type B and type C crystallinity shifted to the smaller degree side than that of the original starch, suggesting the expansion of the interlayered distance by the invasion of water molecules. The invasion of water molecules into the structure expanded the lattice spacing of the cavity from 1.56 nm to 1.61 nm, which corresponded to about 1/6 of the diameter of a water molecule.

**Keywords:** type B crystallinity, expansion by water, sago starch, X-ray diffraction

## Introduction

Starch granules consist of two types of higher molecular weights, amylose and amylopectin, which have a crystalline structure (Nikuni, 1956; Wang et al., 1988; Gallant et al., 1997; Blazek and Gilbert, 2011). X-ray diffraction (XRD) analysis (Buerger, 1937) has been developed and has provided the information on crystalline materials, the distance of the reflection surface, the size of the crystalline structure, and so forth. Katz (1930) and Katz and van Itallie (1930) studied the physical chemistry of starch and classified starches into some groups, essentially type A and type B, based on the XRD patterns. Nikuni et al. (1948) also tried to analyze starches using XRD. The structural elements of amylose are double helices, which are packed right-handed and parallel-stranded into type B (Wu and Sarko, 1978a) and type A (Wu and Sarko, 1978b) starches. The amylose helices of type B and type A are packed to be antiparallel. Eisenhaber and Schultz (1992) subsequently revealed the left-handed and antiparallel helices of amylose using a Monte Carlo simulation. Based on the results proposed by Imberty (1988) and Imberty and Perez (1988), amylose is probably left-

handed and parallel. The central channel (cavity) in the amylose of the type B starch, surrounded by six double helices, is filled with water. On the other hand, type A is quite similar to type B except that the central channel is occupied by another double helix, making the packing closer (monoclinic system). Hizukuri and Nikuni (1957a, b) suggested that the type C starch provided the X-ray diffraction pattern of the mixture of type A and type B. The temperature during the germination of soybean seedlings (Hizukuri et al., 1961) and the soil temperature during sweet potato growth (Nikuni et al., 1963) made the change in the crystalline structure.

In addition, Hizukuri et al. (1983) revealed that the amylopectin of the type A starch had a shorter chain length than that of type B based on the data of 27 starch samples (16 type A, 7 type B, and 4 type C). The amylopectin molecular weight of type A and type B was  $1.2 \times 10^8$  Da and  $2.8 \times 10^8$  Da, respectively (Jane et al., 2003). Hizukuri (1985) found the relation of type A, type B, and type C starches to the weight-average chain lengths of amylopectin and concluded that the chain lengths of the amylopectin of type A, type B, and type C starches were in the ranges of

23–29 Degrees of polymerization (DP), 30–44 DP, and 26–29 DP, respectively. The schematic model for the arrangement of amylopectin in potato starch was proposed by Oostergetel and van Bruggen (1993). They insisted that the 5 nm crystalline lamella contains double helical linear segments. The crystallites are packed into a continuous network consisting of left-handed helices packed in a tetragonal array. Four amylopectin molecules are projected into one of the helices. Meanwhile, Kozlov et al. (2007) proposed schematic amylose localization in the amylopectin clusters of potato starch by the joint analysis of the small-angle X-ray scattering and differential scanning calorimetry technique.

The physicochemical, rheological, and structural characteristics of sago starch have been studied from the perspectives of home economics, cookery science, and the starch industry (Takahashi et al., 1981; Yatsugi, 1986; Takahashi, 1986; Oya and Takahashi, 1987a; Oya and Takahashi, 1987b; Oya et al., 1990; Takahashi and Hirao, 1992; Takahashi and Hirao, 1994; Fasihuddin and Williams, 1999; Fasihuddin et al., 1999; Hamanishi et al., 2000b; Sopade and Kiaka, 2001; Hamanishi et al., 2002; Hirao et al., 2002; Hirao and Igarashi, 2002; Md Zaidul et al., 2003; Konuma et al., 2012; Odeku, 2013; Singh and Nath, 2013). The characteristics of native, modified, and resistant starches from sago have been elucidated by Takeda et al. (1989), Gernat et al. (1990, 1993), Cui and Oates (1999), Wattanachant et al. (2002), Leong et al. (2007), Karim et al. (2008), and Yaacob et al. (2001). Polnaya et al. (2013) studied the effects of phosphorylation and cross-linking on the sago starch structure, and Pukkahuta and Varavinit (2007) found the structure transformation by heat moisture and osmotic pressure. In recent years, Ratnam et al. (2003) reported the production of ethanol from sago starch. Ismal and Zaaba (2012) made plastic film from sago starch, and Watanabe and Ohmi (1997) reported the thermoplasticization of acetylated sago residue after the extraction of starch.

The XRD for analyzing the structure of sago starch

was used by Takahashi et al. (1981), Kawabata et al. (1984), and Yatsugi (1986) to compare to those of corn, potato, mung bean, cassava, and arrowroot. Yatsugi (1986) reported that four sago starch samples collected from East and West Malaysia did not show the difference in the XRD patterns, although four sago starch samples remarkably indicated the difference in the first and fourth peaks of XRD patterns.

Starch scientists should look at the clear peak at around  $5^\circ$  of Cu  $K\alpha$  among several sharp peaks, because the lower peak angle has scarce peak piling. The reflection peaks at a lower angle hold the important information on starch structure.

The XRD pattern of sago starch at various portions of the trunk at different growth stages (9 to 14.5 years after transplanting) and the relative crystallinity of sago starch corrected by the internal standard of  $\text{CaF}_2$ , ranging from  $22$  to  $40^\circ$ , have been reported (Hamanishi et al., 1999; Hamanishi et al., 2000). Fasihuddin et al. (1999), Okazaki et al. (2010), and Yaacob et al. (2011) also reported that sago starch showed the peak at around  $5^\circ$  of X-ray diffraction.

Recently, Polnaya et al. (2013) revealed that native sago starch from Indonesia was characterized by a weak diffraction peak at  $2\theta = 5.67^\circ$  and broad peaks at  $2\theta = 15.30^\circ$ ,  $17.12^\circ$ ,  $18.08^\circ$ , and  $23.46^\circ$ , which indicated the type C starch. Uthumporn et al. (2014) also showed that the XRD patterns of sago starch at different growth stages from Malaysia were type C. However, the XRD analysis of sago starch was not sufficient to show an understanding of the sago starch structure during wet and dry processes.

The structural changes of starch accompanied by the addition of water molecules have been studied using  $^1\text{H}$  NMR (Mousseri et al., 1974; Botlan et al., 1998; Tang et al., 2000). Generally, however, the sensitivity of NMR for the behavior of water molecules is still low compared to other analytical methods. The determination of water molecule dynamics in starch by  $^1\text{H}$  NMR, therefore, faced the limitation derived from the sensitivity of NMR, whereas X-ray diffraction is a useful tool for

analyzing the expansion and shrinkage of the starch structure at the nanometer level with the invasion of water molecules. Hizukuri and Nikuni (1957a) reported that the full width at half maximum became larger upon increasing the water content from 10.5 to 47.5%, which indicated a decrease in the crystallinity index (Katsumi et al., 2014). However, the XRD analysis of the type C starch (for example, sago starch) was not sufficient to understand the flexibility of the starch structure during wet and dry processes.

In this study, we attempt to expand and shrink the channel (cavity) space surrounding the six double helices of type B and type C starches using water molecules through the peak shifting at around 5 to 6° of X-ray diffraction.

## Materials and Methods

### 1. Starch samples

Sago starches were collected from Leyte in the Philippines in 2006. The sago starch from the log was extracted by macerating the pith in water in a blender. The starch was separated from the pulp by squeezing the pulp in a cotton cloth and then air-dried. The potato starch was from Miyazawa Pharmaceutical Co., Ltd.

### 2. Addition of water to starch samples

About 2.0 g of the samples was added to 1.5 mL Milli-Q water and heated in a water bath for 10 min at 40°C. The moisture content of starch samples was determined using the semi-micro balance (Sartorius CPA224S) before and after the addition of water to starch samples in the small glass container.

### 3. XRD and curve fitting

The X-ray identification was carried out using an X-ray diffractometer (Mini Flex, Rigaku) operating at 30 kV and 15 mA. Copper (Cu) K $\alpha$  radiation was

used with a nickel filter. The scanning speed was recorded at 2° min<sup>-1</sup>. The operation angle was 3 to 45° for 2 $\theta$ . XRD data were treated with the Lorentz polarization factor correction, the scattering factor correction, and the absorption factor correction (Rigaku, 1962). After those corrections, the smoothing treatment was performed using a weighted mean method (smoothing number: 11). The background subtraction was done using the Sonneveld–Visser method (peak width: 0.25; threshold intensity: 0.01), and the curve fitting was introduced in the diffraction angle at 4 to 7°. Curve fitting was carried out with Gauss function using PeakFit version 4.12 (SeaSolve Software Inc.). An X-ray diffraction crystalline index (CI) for powder was carried out by height and the full width at half maximum (FWHM) of the third b (3b) and fourth a (4a) peaks after the correction and wave-form separation (Katsumi et al., 2014).

## Results

### 1. XRD pattern of sago starch

The water content of native starch samples was 14% for sago and 16% for potato. The wide-angle XRD patterns of native sago and potato starch samples are shown in Fig. 1. The peaks at around 5.6° (1.58 nm), the double peak at 17 (0.52 nm) and 18°

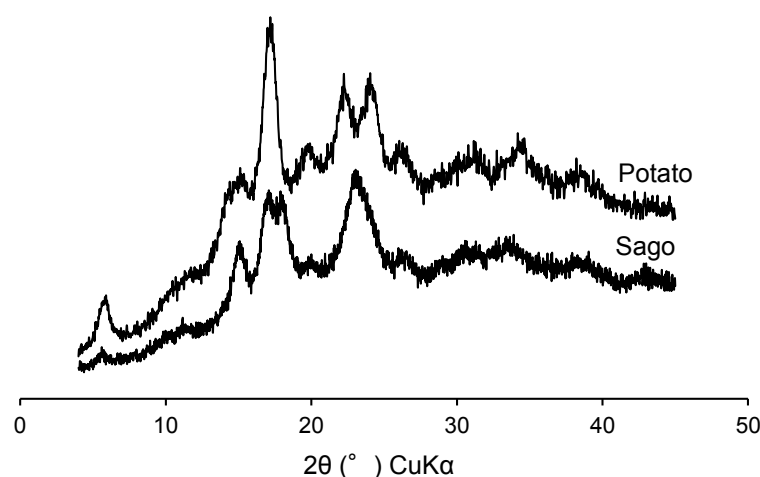


Fig. 1. X-ray diffraction pattern of starch samples

(0.49 nm), and the peak at  $23^\circ$  (0.39 nm) of sago starch showed type C crystallinity, which held the diffraction characteristics of a type A and type B mixture (Hizukuri and Nikuni, 1957b). This coincided with the result of Katsumi et al. (2014). Meanwhile, potato starch provided the peak at around  $5.6^\circ$  and  $17^\circ$  and the double peak at  $22^\circ$  (0.40 nm) and  $24^\circ$  (0.37 nm), which showed the characteristics of type B crystallinity (Cairns et al., 1997) (Fig. 1). Potato starch clearly showed the peak at around  $5.6^\circ$ , which was derived from the cavity diameter of the structure.

The CI value determined by the peak of 3b and 4a was 609 for native sago starch (type C) and 879 for native potato starch (type B) (Katsumi et al., 2014). There was no significant difference in CI values before and after the presence of additional water in this study.

## 2. Expansion of sago starch lattice by water molecules

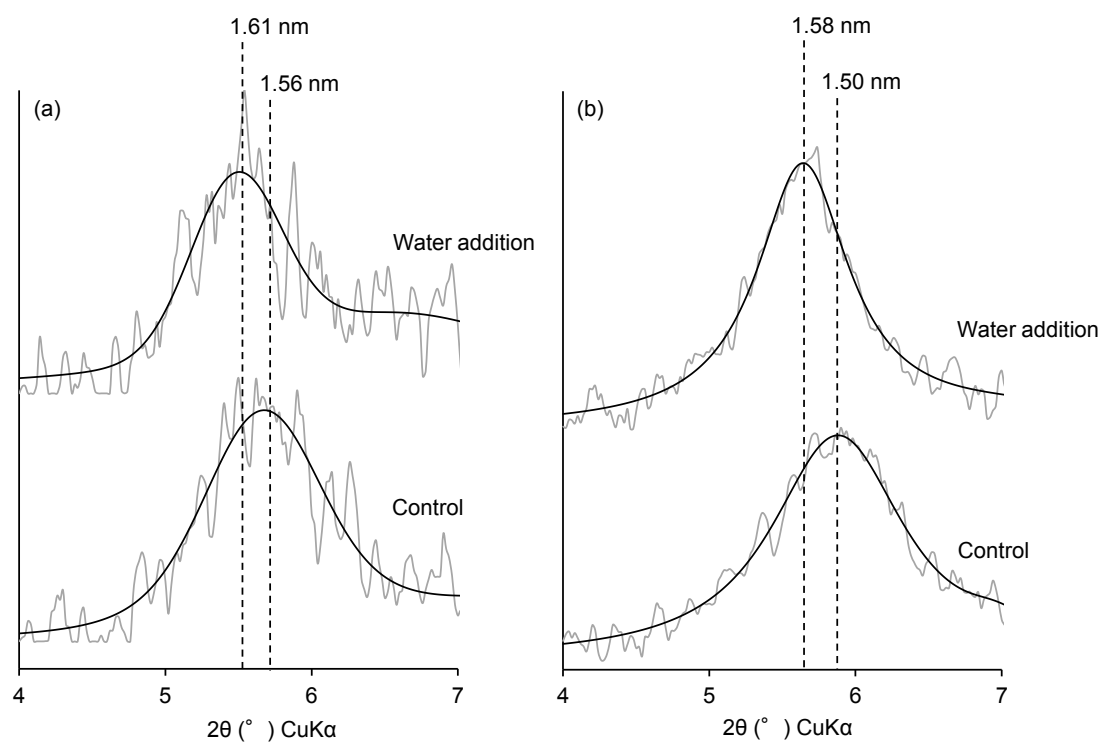
The XRD peak at about 5 to  $6^\circ$  for  $2\theta$  of water-added starch shifted to a smaller degree than that of

the original starch, suggesting the expansion of the distance by the invasion of water molecules (Fig. 2). Invasion of the water molecules into the structure of sago starch (Fig. 2 (a)) expanded the lattice spacing from 1.56 nm to 1.61 nm, which corresponded to about 1/6 of the diameter of a water molecule. Potato starch (Fig. 2 (b)) expanded the lattice spacing from 1.50 nm to 1.58 nm by the invasion of water molecules inside the starch structure. Potato starch (type B) showed larger expansion than did sago starch (type C), as much as 1.6 times.

## Discussion

### 1. Behavior of water molecule in starch granule

The water saturation in the cavity of the type B starch gave rise to a 1.58-nm interlayer distance in the double helical structure (Blazek and Gilbert, 2011), which reached the full interlayer distance between the double helix structure. Sago starch shows the type A starch mixed with the type B starch as an accessory.



**Fig. 2.** X-ray diffraction pattern of sago (a) and potato (b) starches  
The curve fitting was carried out with a Gauss function using PeakFit Version 4.12  
(Sea Solve Software Inc., Framingham, MA, USA).

Even though sago starch gave a small amount of the type B starch, the expansion of the interlayer distance by the invasion of water molecules occurred.

## 2. Flexibility of the starch structure with water molecule invasion

A recent review of the starch structure focused the macromolecular conformation of starch. Kozlov et al. (2007) revealed that amylose chains were located in the amorphous lamella of amylopectin. Oostergetel and van Bruggen (1993) proposed the three dimensions of amylopectin molecules and illustrated that the crystalline layers containing the double helical linear segments in the amylopectin molecules formed a continuous network consisting of the left-handed helices packed in a tetragonal array. Imberty and Perez (1988) revealed the presence of 36 water molecules per unit in the amylose structure of the type B starch, and the addition of water to starch in this study caused the expansion of the interlayer distance from 1.56 to 1.61 nm, which corresponded to about 1/6 of the diameter of a water molecule. This indicates the flexibility of the type C sago starch structure with regard to adding water molecules as well as that of the type B potato starch.

## Conclusion

The XRD shows that sago starch has type C crystallinity (containing type A and type B) and exhibits the expansion of the cavity by the invasion of water molecules. These structural characteristics of sago starch provide appropriate water-holding properties as dusting flour for noodles (Hirao, 2015) on the basis of the invasion of water molecules into the cavity of the type B and type C structures of starch.

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