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Complexation of allyl isothiocyanate by α- and β-cyclodextrin and its controlled release characteristics

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Abstract

The complexation of allyl isothiocyanate (AITC) by α- and β-cyclodextrin (CD) and its controlled release characteristics were investigated in this paper. Almost 100% of inclusion capacity was achieved at the AITC/CD molar ratio of 2:1 using a coprecipitation method, combined with water washing. The controlled release behaviour of the incorporated AITC was significantly affected by relative humidity (RH) and the type of CD. The release was accelerated with increased RH, and the release rate of AITC in the α-CD-AITC complex was much slower than that in the β-CD-AITC complex. Subsequent analysis with GC demonstrated that the headspace concentration of AITC over β-CD-AITC complex in a hermetical system was significantly dependent on RH, and could finally be at a different equilibrium level at various storage times.

Keywords: Cyclodextrin; Allyl isothiocyanate; Inclusion complex; Controlled release

1. Introduction

Allyl isothiocyanate (AITC, CH_2=$CHCH_2N$=C=S), the major pungent compound in plants belonging to the Cruciferae family, such as cabbage, horseradish and mustard, has been shown to have strong antimicrobial activity in its vapour form (Delaquis & Sholberg, 1997). It was reported that AITC could kill fungal and bacterial pathogens on plant seeds, fresh produce, bread, meat and cheese (Delaquis & Mazza, 1995; Lin, Kim, Du, & Wei, 2000; Nadarajah, Han, & Holley, 2005; Nielsen & Rios, 2000; Park, Taormina, & Beuchat, 2000; Suhr & Nielsen, 2005). The minimum inhibitory concentrations of AITC toward bacteria, yeasts and molds were about 34–110, 16–37 and 16–62 ng/ml, respectively (Isshiki, Tokuoka, Mori, & Chiba, 1992), which suggested that AITC might be a potential natural antimicrobial agent for food preservation. However, the application of AITC in food systems is limited due to its volatility and strong odour which significantly affect the taste of food (Chacon, Buffo, & Holley, 2006). Its application in food packaging systems is also limited by its existence as an oil form which is not compatible with major food packaging materials.

Cyclodextrins (CDs) are cyclic oligosaccharides with the characteristic molecular structure of a hydrophilic exterior and hydrophobic central cavity, capable of forming inclusion complexes (IC) with many organic compounds by incorporating them into the CD cavities in the presence of water (Hedges, 1998). The driving forces have been attributed to various factors, including Van der Waals force, hydrophobic effect and dipole–dipole interactions (Liu, Song, Li, & Guo, 2001). As a result of complex formation, the physicochemical properties of the guests can change significantly, and this promises an extensive industrial application of CDs, which include improvement of

Abbreviations: AITC, allyl isothiocyanate; CDs, cyclodextrins; IC, inclusion complex; RH, relative humidity; α-CD-AITC, the AITC inclusion complex with α-CD; β-CD-AITC, the AITC inclusion complex with β-CD.

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solubility, process aids, stabilization, reduction of volatility and masking effects of the guest (Hashimoto, 2002; Hedges, 1998).

AITC, being a small molecular weight organic compound, can be incorporated in α- and β-CD (Ohta, Takatani, & Kawakishi, 1999). As a result of the complexation, a controlled release of AITC from the complexes may be achieved, which is of great benefit in masking the strong odour, prolonging the antimicrobial time and enhancing the antimicrobial effect of AITC. In recent years, there have been some reports of the application of CDs in packaging material (Hashimoto, 2002; Lu et al., 2001). It seems possible for AITC incorporated in CDs to be applied in antimicrobial food packaging systems. However, to the best of our knowledge, there is a limited literature on the complexation of AITC by CDs and its controlled release, even though several studies have dealt with the effect of CDs on AITC in an aqueous solution (Ohta, Matsui, Osawa, & Kawakishi, 2004; Ohta et al., 1999, Ohta, Takatani, & Kawakishi, 2004). Therefore, the aim of this paper was to investigate the inclusion of AITC in α- and β-CD by a coprecipitation method and to study the controlled release behaviour of the included AITC at various relative humidites (RH).

2. Materials and methods

2.1. Materials

AITC (>98% GC purity) was purchased from Fluka. Chem. Co. (Germany). α- and β-CD (99% purity) were purchased from Shanghai Seebio Biotechnology Inc. (China). All other reagents used were of analytical grade.

2.2. Preparation of AITC inclusion complexes with α- and β-cyclodextrin (α- and β-CD-AITC)

β-CD-AITC complex was prepared, using a coprecipitation method described by Bhandari, D'arcy, and Bich (1998) with minor modifications. β-CD (5 g) was dissolved in 150 ml of distilled water at 60 °C on a hot plate. After cooling the β-CD solution to 40 °C, AITC in ethanol (1:1, v/v) was slowly added to the solution with continuous agitation, to give a molar ratio of AITC/β-CD of 0.4–2.4. The vessel was sealed and kept under stirring for 3 h, and then the resulting slurry was refrigerated overnight at 4 °C. The cold precipitate was recovered by vacuum-filtration (and washed or not with 15 ml distilled water), and dried in a vacuum oven (ZK82A, Shanghai Laboratory Instrument Works Co., Ltd., Shanghai, China) at 75 °C for 24 h. The final dry complex powders were stored in an airtight glass desiccator at room temperature.

The above-mentioned method was also used for the preparation of α-CD-AITC inclusion complex, except that α-CD (5 g) was dissolved in 35 ml of distilled water at 60 °C on a hot plate and the molar ratio of AITC/α-CD was 2:1.

The total recovery was calculated according to the following Eq. (1):

$$\text{Total recovery } (\%) = \frac{\text{Recovered powder}}{\text{Initial (CD + AITC)}} \times 100 \qquad (1)$$

2.3. Surface AITC determination

Surface AITC of the complex powder was determined by a washing method, according to Padukka, Bhandari, and D'Arcy (2000) with some variations. The dry complex (0.10 g) was added to 10 ml of hexane and the mixture was gently shaken for 20 min at room temperature and the solvent phase containing AITC was separated from the test tube by decantation. The residue was further washed for 10 min with 10 ml of hexane and the procedure was repeated once. The volume (V) of the combined extracts was measured, and the UV absorbance at 248 nm was recorded by a spectrophotometer (UV-1700, Shimadzu Corp., Japan). The concentration of AITC in the extracts (C) was assessed by plotting the calibration curve of AITC standards (from 0.01 to 0.07 μl/ml). The equation of the calibration curve was $C = 0.0959\ A_{248}$, and the coefficient, R^2, was 1.0, where C is the AITC content in μl/ml of extracts and A_{248} is the absorbance at 248 nm. The total content of AITC extracted was calculated using the product of V and C. The determination was carried out in triplicate for one sample.

2.4. Total AITC determination

The total content of AITC in complex powder was measured according to a solvent (hexane) extraction method described by Padukka et al. (2000) with some modifications. Complex powder (0.10 g) was weighed into a 50 ml flask and mixed with 5 ml of distilled water and 7 ml of hexane. The flask was connected to an upright glass condenser cooled by tap water. Then the mixture was heated in a water bath at 85 °C for 20 min with intermittent shaking. On heating, a glass lid was attached to the top of the condenser, to avoid the loss of AITC. After the first extraction was finished, the flask was cooled to room temperature and the inner wall of the condenser was washed with 2–3 ml of hexane in order to collect the maximum amount of AITC. After that, the upper hexane, containing AITC, was separated by pipette. The extraction was repeated three times. Since only a very minute amount of AITC was detected by UV absorbance in the fourth extract, four extractions were sufficient to extract all the AITC. Finally, the volume (V) and the absorbance at 248 nm (A_{248}) of the combined hexane extracts were measured. The concentration of AITC (C) of the extracts was assessed by the above-mentioned calibration curve of AITC standards. The final amount of the total AITC of complex powder was calculated using the product of V and C. The determination was carried out in triplicate for one sample. The CD recovery and inclusion efficiency of CD were calculated according to the following Eqs. (2) and (3), respectively:

$$\text{CD recovery } (\%) = \frac{\text{Recovery powder} - \text{Total AITC}}{\text{Initial CD}} \times 100 \quad (2)$$

$$\text{Inclusion efficiency of CD } (\%) = \frac{\text{Total AITC} - \text{Surface AITC}}{\text{The theoretical maximum included AITC}} \times 100 \quad (3)$$

2.5. Release characteristic of AITC from the complex powder

α- or β-CD-AITC complex powders were weighed and spread flatly on watch glasses(Φ 60 mm) and placed in three uniform desiccators (Φ 400 mm) at 50%, 75% and 98% of RH, which were controlled by saturated salt solutions of $Mg(NO_4)_2$, NaCl and K_2SO_4, respectively (Rehmann, Yoshii, & Furuta, 2003; Shiga et al., 2001); each desiccator contained two watch glasses with one gramme of α- and β-CD-AITC powder. After that, they were stored for six days at 28 °C. At 12 h intervals, the lids of desiccators were opened and humid air of the same relative humidity was blown into the desiccators to purge the gas. At 24 h intervals, about 0.1 g samples of α- and β-CD-AITC were removed from the desiccators and the residual amounts of AITC in the powders were measured by the method of total AITC described above, and then the retention of AITC in the powder was expressed as the ratio of the initial AITC content.

2.6. Headspace concentration above β-CD-AITC powder at various RH by GC

Three glass vials (ca.10 ml) with 0.60 g of β-CD-AITC powder were separately placed in three 250 ml glass jars at 50%, 75% and 98% of RH controlled by saturated salt solutions (20 ml) at room temperature. The jars had their lids processed to make a hole (Φ 5 mm) sealed with a silicone rubber stopper. Great care was taken to avoid the salt solution flowing into vials to wet the complex powder. At particular intervals, one millilitre of headspace gas was collected through the stopper with a gastight syringe (1001, TLL with slots, Hamilton Co., Bonaduz, Switzerland) and injected into a gas chromatograph with a flame-ionization detector (GC-2010, Shimadzu Corp.) for analysis. The syringe was heated in an oven prior to headspace sampling to drive out the remainder of the AITC vapour in the syringe barrel. Pure AITC was used as a control. A calibration curve for GC analysis was prepared by injecting AITC standards (in hexane) of known concentration (Lim & Tung, 1997).

A capillary DB-1 column (J&W Scientific, Folsom, CA, USA) was used (30 m × 0.53 mm i.d., film thickness: 1.5 μm). The operating conditions were as follows: oven temperature programme: from 60 (held for 1 min) to 100 °C at a rate of 10 °C/min, then held at 100 °C for 2 min. The carrier gas was N_2 at a flow rate of 10 ml/min. The temperature of injection port was 200 °C and that of FID was 250 °C. Hydrogen and air flow rates were 47 ml/min and 400 ml/min, respectively.

3. Results and discussion

3.1. Preparation of α- and β-CD-AITC complexes

The preparation of inclusion complex is widely performed using the coprecipitation method in the laboratory, which has the advantage of easy observation of the complex forming and the guest disappearing during the inclusion (Hedges, 1998). The content of AITC, in this study, was determined using UV colorimetry instead of the GC method used in many previous studies (Isshiki et al., 1992; Ohta et al., 1999, Ohta, Takatani, & Kawakishi, 2004). AITC possesses ultraviolet absorption due to its unsaturated-bond in the molecule. The used AITC (in hexane) solution in this study showed a maximum absorbance at 248 nm by UV scanning analysis. Compared with GC analysis, the UV spectrophotometer is much simpler, fast, and easy to operate, which is suitable to the determination of AITC.

The main factors affecting the form of the inclusion complex are the concentration of CD, the molar ratio of guest to host, the reaction temperature, the addition of ethanol and washing with water. Among these, the molar ratio of the guest to CD was the key factor. As shown in Fig. 1, during the preparation of β-CD-AITC, the content of AITC included in the complex increased with increase of the molar ratio of AITC/CD, and a plateau of 68.7 μl/g IC (73.8 μl/g CD) was obtained at a ratio of 2:1. A similar trend was also observed in the CD recovery (from Fig. 1). The α-CD-AITC inclusion complex was prepared at a AITC/CD molar ratio of 2:1, and the content of included AITC in the complex was 81.7 μl/g IC (89.0 μl/g CD). Ohta et al. (2004) reported that AITC was suitable for incorporation in α- or β-CD in aqueous solution and would form

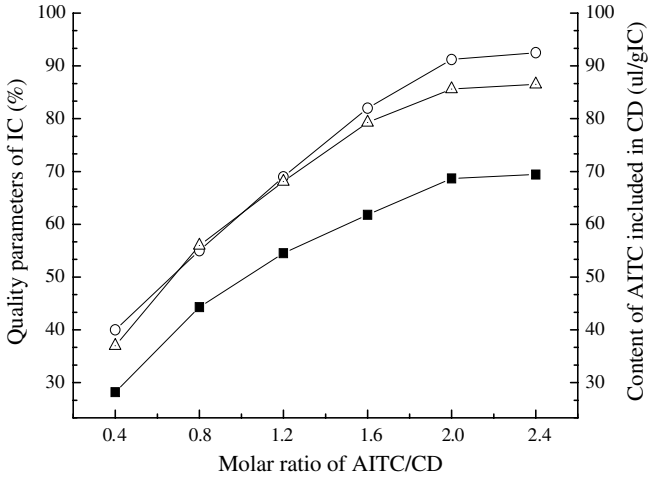

Fig. 1. The effect of the molar ratio of AITC/β-CD on inclusion reaction. (○), CD recovery (%); (△), the inclusion efficiency (%); (■), the content of AITC included in the complex (μl/g IC).

CD-AITC complex in 1:1 molar ratio. Based on that result, the theoretical maximum inclusion contents of AITC were 86.3 μl of AITC for one gramme of β-CD and 101 μl of AITC for one gramme of α-CD. Thereby, in this study, the inclusion efficiencies, represented by the percentage of the complexed CD in total CD, were 85.6% for β-CD-AITC complex powder and 88.4% for α-CD-AITC complex powder.

The effect of washing with water or ethanol on inclusion capacity of the complexes was also investigated (Table 1). Washing with water could improve the inclusion efficiency (almost 100% of the inclusion efficiency of CDs was obtained), whereas the total recovery and the CD recovery were decreased. The improved inclusion capacity of the complexes was considered to be the reason why the free CD was more water-soluble than were the co-crystallized CD complexes, and they could easily be dissolved in water and removed by filtration. Washing with ethanol significantly decreased the loaded content of AITC in the complexes (data not shown), which might be due to the competitive binding with CDs between ethanol and AITC that impelled the dissociation of AITC from the complexes.

3.2. Release characteristic of AITC from the complex powders

It has been reported that the presence of water was essential for the formation of the inclusion complexes between CDs and hydrophobic guests (Yoshii, Furuta, Yasunishi, & Hirano, 1994). In fact, the inclusion reaction was a substitution process of guest for water in the cavity of the CD and vice versa (Rehmann et al., 2003). Obviously, the release of the guest from the CD might be markedly affected by the environmental RH.

As shown in Fig. 2, AITC could be gradually released from the complexes at the given RH, and its release rate was greatly accelerated with the increase of RH. Whether it is α- or β-CD-AITC, the content of AITC in the complexes was quickly reduced at the initial stage of storage, and then the release became very slow, and finally the release very nearly ceased altogether, which demonstrated that the release rate of included AITC was not only dependent on RH but also on the content of AITC in the complexes. The AITC in α-CD-AITC had a much lower release rate than had that in β-CD-AITC. This result indicated that AITC bound more strongly with α-CD than with β-CD, which might be due to the molecule size of AITC being more matched with the cavity of α-CD. This is in accordance with the findings of Ohta et al. (1999).

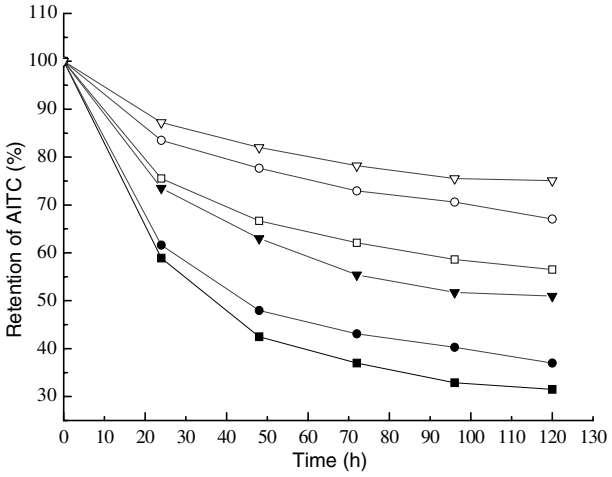

Fig. 2. Release behaviour of AITC from α- and β-CD-AITC complexes under various RH conditions. (▽), α-CD-AITC, 50% RH; (○), α-CD-AITC, 75% RH; (□), α-CD-AITC, 98% RH; (▼), β-CD-AITC, 50% RH; (●), β-CD-AITC, 75% RH; (■), β-CD-AITC, 98% RH.

The Avrami equation (Eq. (4)), which has been recently applied to represent solid–gas and solid–liquid reaction schemes (Shiga et al., 2001), well simulated the retention time curve of flavour in CD complex powder (Rehmann et al., 2003). In this study, the Avrami equation was used to evaluate the release rate constant of AITC under the experimental conditions, as follows:

$$R = \exp[-(kt)^n], \tag{4}$$

where R is the retention (%) of the volatile, t is the release time (s), k is the release rate constant, and n is a parameter representing the release mechanism. $n = 1$ corresponds to the first-order release kinetics and $n = 0.54$ represents the diffusion-limited system (Shiga et al., 2001).

The estimated parameters of the equation are listed in Table 2. All of the n values estimated were near 0.54, which indicated that the release of included AITC in the complexes corresponded to the limited diffusion kinetics system. A higher k value obtained at a higher RH, as displayed in Table 2, further demonstrated the significant effect of RH on the release behaviour of included guest in the complexes.

3.3. Headspace concentration above β-CD-AITC complex at various RHs by GC

The release behaviour of included AITC in the complex in a hermetical system was further investigated by determining the headspace concentrations of AITC above the

Table 1
The quality parameters of the inclusion complexes of AITC with α- and β-CD[a]

CD-AITC	Total AITC (μl/g IC)	Surface AITC (μl/g IC)	Total recovery (%)	CD recovery (%)	Appearance
β-CD-AITC	72.6/84.4[b]	3.9/5.3[b]	83.8/79.5[b]	91.2/85.3[b]	White powder (loose, no odour)
α-CD-AITC	83.4/91.5[b]	1.7/1.1[b]	41.3/38.6[b]	45.2/42.1[b]	White powder (loose, no odour)

[a] The reaction time is 3 h, and the molar ratio of AITC/CD is 2:1.
[b] Washing with water.

Table 2
Values of k and n on the release of AITC from α- and β-CD-AITC at various RHs

RH (%)	β-CD-AITC			α-CD-AITC		
	n	$k \times 10^{-6}$ (s^{-1})	R^2	n	$k \times 10^{-6}$ (s^{-1})	R^2
50	0.4799	1.1383	0.9993	0.3788	0.0842	0.9972
75	0.5249	2.6900	0.9987	0.4102	0.2163	0.9988
98	0.5440	3.6166	0.9988	0.4588	0.7426	0.9997

β-CD-AITC complex with GC. When pure AITC (50 μl) was introduced to a 250 ml glass jar and equilibrated for 1 h at room temperature, 896 ng/ml of AITC headspace concentration was obtained. Compared with that of pure AITC, the headspace concentration of AITC over the β-CD-AITC complex was very low and was affected by RH. As shown in Fig. 3, a dynamic equilibrium was reached at 75 h of storage time, at 98% RH, with an AITC headspace value of 386 ng/ml, and a similar equilibrium was reached at 90 h of storage time at 75% RH with an AITC headspace value of 288 ng/ml. In the case of 50% RH, only an AITC headspace concentration of 61 ng/ml was obtained at 105 h of storage time and the release proceeded. These results demonstrated that both the magnitude of equilibrium headspace concentration of AITC and the time for the equilibrium were dependent on the RH.

Although a reaction model (Eq. (5)) has been employed to analyze the inclusion process of CD in the micro-aqueous system (Yoshii et al., 1994), Kant, Linforth, Hort, and Taylor (2004) reported that the presence of β-CD created a reservoir for binding some volatiles in their solution and the binding was readily reversible. This research suggested that the interaction of AITC with CD was also readily reversible and a dynamic equilibrium process existed during the release of the included AITC from the complexes. According to the results mentioned above, a modified reaction model (Eq. (6)) was developed to elucidate the release mechanism of AITC from the inclusion complex.

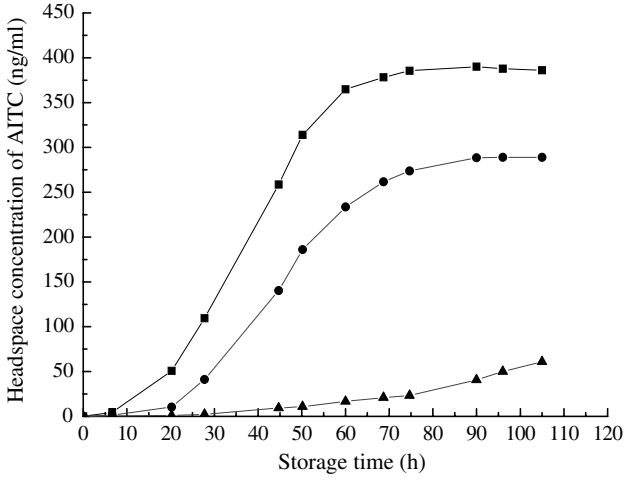

Fig. 3. The headspace concentration of AITC over the β-CD-AITC complex by GC under various RH conditions. (▲), β-CD-AITC, 50% RH; (●), β-CD-AITC, 75% RH; (■), β-CD-AITC, 98% RH.

$$\mathrm{CD} - n\mathrm{H_2O} + \mathrm{Guest} \xrightarrow{k} \mathrm{CD} - \mathrm{Guest} + n\mathrm{H_2O} \quad (5)$$

$$\mathrm{CD} - \mathrm{AITC} + n\mathrm{H_2O} \underset{k2}{\overset{k1}{\rightleftarrows}} \mathrm{CD} - n\mathrm{H_2O} + \mathrm{AITC} \quad (6)$$

Based on Eq. (6), the release behaviour of the included AITC in CDs in a special system, to some extent, abides by the dynamic equilibrium mode of the gas–solid reaction, in which both RH and the content of included AITC are crucial factors. In the presence of an AITC absorbent, the original equilibrium concentration is decreased and a new equilibrium concentration is derived from the controlled release of the included AITC.

4. Conclusion

The complexes of AITC with CDs were easily formed. Almost 100% of inclusion capacity was achieved at the AITC/CD molar ratio of 2:1, using a coprecipitation method combined with water washing. The complexes obtained possessed the controlled release characteristics which were significantly affected by RH. GC analysis further revealed that the interaction of AITC with CDs was readily reversible and a dynamic equilibrium process existed during the release of the included AITC from the complexes.

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