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Inclusion Complex of Plai Oil and β-Cyclodextrin

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Keywords: Plai *(Zingiber cassumunar*) oil; β-cyclodextrin; inclusion complex.

Abstract. Inclusion complex of β-cyclodextrin (β-CD) and Plai *(Zingiber cassumunar*) oil was prepared using a simple co-precipitation method at β-CD to Plai oil in different ratios. The inclusion complexes were characterized using Fourier transform-infrared spectroscopy (FT-IR) and differential scanning calorimetry (DSC). The FT-IR absorption bands of inclusion complex at 3600-3200 cm^{-1} were broader and shifted toward lower frequencies compared with that of pure β-CD (3359 cm^{-1}). DSC of the inclusion complexes showed two endothermic peaks shifted to lower temperatures (90-100°C and 295-300°C) compared to that of β-CD. The different physicochemical characteristic could be an indication of an embedded guest molecule in the β-CD cavities in the inclusion complex preparation.

Introduction

In the latest year, cyclodextrin (CD) complexation has been effectively used to improve the solubility and stability of a number of poorly water-soluble compounds. CD is cyclic oligosaccharides made up of six to twelve α-D-glucopyranose monomers, which are linked by α(1→4) glycosidic bond to form a cylindrical structure [1]. CD has a hydrophilic rim and a lipophilic interior [2] and with this specific structure, it possesses an ability to form host-guest inclusion complexes with many lipophilic compounds [3]. This property has received extensive attention in the pharmaceuticals and cosmetics to improve the water solubility, stability, dissolution and release rates of various active molecules [4,5]. Complexation of oils [6,7] with β-CD has been studied to protect the oils against oxidation, heat and light degradation.

Zingiber cassumunar Roxb., (also called "Plai" in Thailand) is a medicinal plant widely cultivated in Thailand and tropical Asia. It is frequently used as an ingredient in marketed phytomedicines [8,9]. The rhizome of *Z. cassumunar* has an anti-inflammatory activity. It has been the source of Thai traditional herbal remedies and extracts for topical application to alleviate inflammation. The chemical composition of the essential oil and non-volatile components from the rhizome have been reported to be (E)-4-(3',4'-dimethoxyphenyl)but-3-en-2-ol and (E)-1-(3',4'-dimethoxyphenyl) butadiene. Topical drug cream with Plai oil is used for treatment of traumatic inflammations, sprains, muscular pain, joint inflammations and related disorders. Pla oil is also used in cosmetic and spa products with the maximum 1% in the formulation [10]. Due to its oil soluble nature, it is generally incorporated in emulsion or oil-based cosmetics. In order to include Plai oil in water-base, appropriated delivery matrix is needed. Cyclodextrin (CD) complexation is one method to deliver oil-soluble into water base system. In this work, inclusion complex of Plai oil-β-CD was prepared using a simple co-precipitation method. The inclusion complexes were characterized using Fourier transform-infrared spectroscopy (FT-IR) and differential scanning calorimetry (DSC).

Materials and Methods

Preparation of Plai oil-β-CD inclusion complex

The complex of Plai oil and β-CD was prepared by using a co-precipitation method. β-CD 5.0 g (±0.01) was dissolved in 100 ml of an anhydrous ethanol to deionized water (1:2 v/v) mixture and maintained at 60 °C on a hot plate stirrer (C-MAG HS 7 Digital IKAMAG® Hot Plate Stirrer, IKA-

US). A portion of 0.5, 1 and 2 g of Plai oil was then slowly added to the solution with continuous agitation. The resultant mixture was treated by ultrasonic bath at 35W (Grant XUBA3 Ultrasonic, Grant In-struments-UK) for 4 h. The final solution was maintained overnight at 4 °C. The cold precipitated Plai oil-β-CD complex was recovered by vacuum filtration. The precipitate was washed twice with 50% ethanol solution to remove Plai oil on the surface of β-CD and dried in an oven at 50 °C until the constant weight is obtained.

Fourier transform infrared spectroscopy (FT-IR)

The FT-IR spectra of Plai oil, β-CD and the inclusion complexes were collected between 4000 and 400 cm^{-1} on a FT-IR spectrophotometer (Perkin Elmer/FTIR Spectrum GX).

Differential scanning calorimetry (DSC)

DSC analysis was carried out for Plai oil, β-CD, and the inclusion complex with a differential scanning calorimeter (Mettler-Toledo DSC822, Milan, Italy). Each sample (3-5 mg) was heated in a crimped aluminium pan at a scanning rate of 10 °C/min between 20 and 400 °C temperature range under a nitrogen flow of 50 ml/min. An empty pan sealed in the same way was used as reference. Reproducibility was checked by running the sample in triplicate.

Results and Discussion

Preparation of Plai oil -β-CD inclusion complex

The inclusion complex of Plai oil and β-CD was prepared by using a co-precipitation method. The aqueous ethanol of β-CD was mixed with portion of 0.5, 1 and 2 g of Plai oil. The complexes are white, fine powder, Fig. 1, with characteristic Plai odor. The total recovery was calculated from the recovered powder over the combined amount of Plai oil and β-CD. The total recovery is 60.0±13.2%, 77.3±2.8% and 68.9±2.1% when use 0.5, 1 and 2 g of Plai oil in the preparation, respectively.

Figure 1. The appearance of Plai oil-β-CD inclusion complexes

Determination of Plai oil in the inclusion complex

Plai oil (100 mg) was dissolved into anhydrous ethanol (50 ml), then 0.5, 1.0, 2.0, 3.0, 4.0 and 5.0 ml solution were taken out and made up to 10 ml with ethanol, respectively. These samples were analyzed by a UV–Vis spectrophotometer (Biochroms Libra S22, Biochrom Ltd, UK) monitoring the absorbance at 280 nm. The concentration (x) and the absorbance (y) of Plai oil had a good relationship with the regression equation of $Y=9.6154x$, $R^2=0.9997$. A 30 mg of the inclusion complex sample and 30 ml of ethanol were mixed and bathed in an ultrasonic wave cleaner. Plai oil was extracted by ethanol from the inclusion complex for 20 min in the ultrasonic condition. The supernatant containing Plai oil was obtained by centrifugation at 3000 rpm for 10 min. The content of Plai oil in ethanol was determined using UV–Vis spectrophotometer at 280 nm by the calibration curve of Plai oil. The content of Plai oil in the complex was 6.76, 11.09 and 18.18%, when use 0.5, 1 and 2 g of Plai oil in the preparation, respectively.

Fourier transform infrared spectroscopy (FT-IR)

The FT-IR spectra of Plai oil, β-CD and the inclusion complex were collected between 4000 and 400 cm^{-1} on a FT-IR spectrophotometer (Perkin Elmer/FTIR Spectrum GX). The results are shown in Fig. 2 and Fig. 3. The FT-IR spectrum of β-CD exhibited its characteristic peaks at 3359 cm^{-1} (broad, O-H group), 2926 cm^{-1} (C-H stretch), 1642 cm^{-1} (H-O-H bending), 1155 cm^{-1} (C-O stretch)

and 1032 cm^{-1} (C-O-C stretch) [11,12]. The IR spectra of the inclusion complexes are relatively similar to that of β-CD, however, absorption bands of inclusion complex at 3600-3200 cm^{-1} were broader and shifted toward lower frequencies compared with that of β-CD. Moreover, the H-O-H bending at 1642 cm^{-1} also exhibited a shifted toward higher frequencies when the amount of Plai oil in the preparation increased.

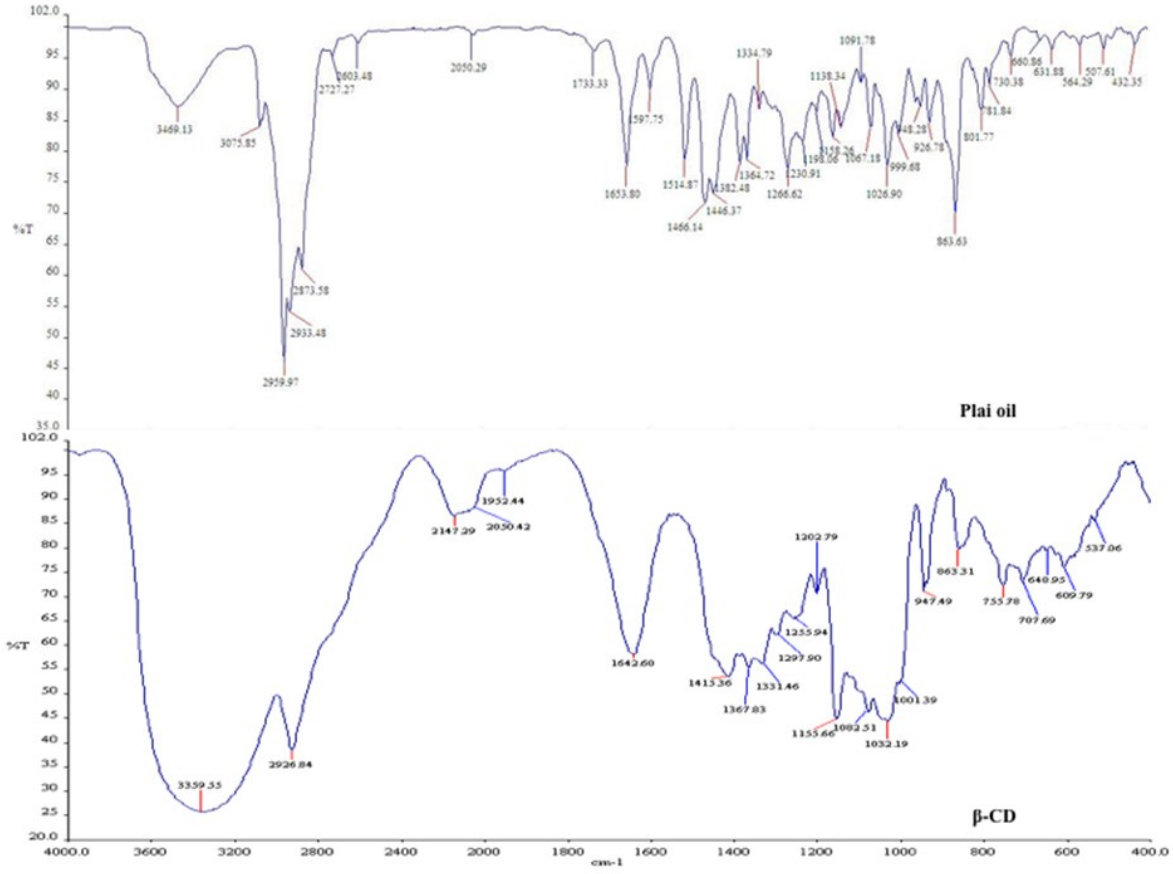

Figure 2. FT-IR spectra of Plai oil and β-CD

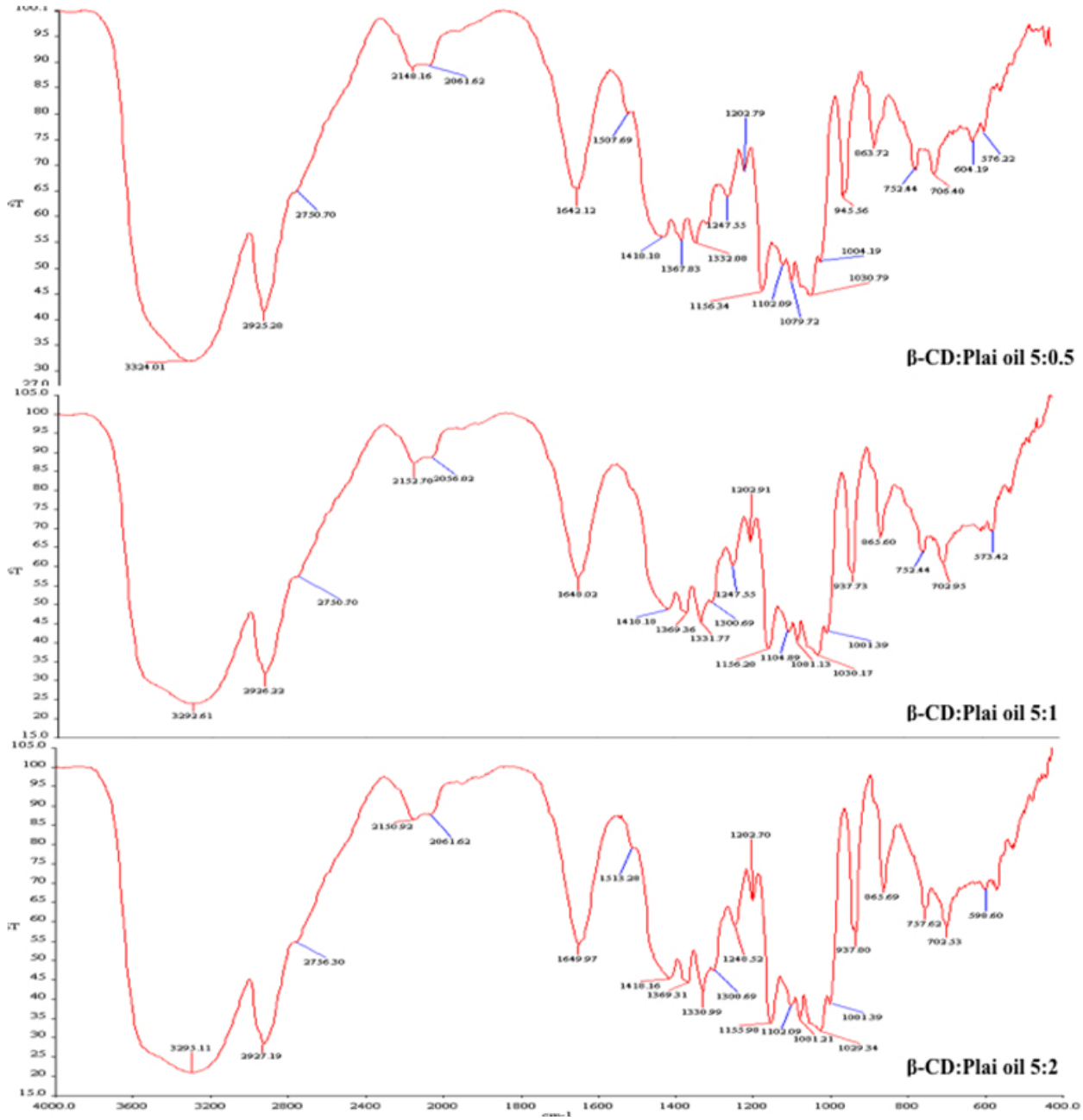

Figure 3. FT-IR spectra of the Plai oil-β-CD inclusion complexes

Differential scanning calorimetry (DSC)

The DSC results of β-CD, Plai oil, and the inclusion complex are presented in Fig. 4 and 5. The DSC curve of β-CD showed an endothermic peak at 136.5 °C, which may due to the elimination of water. There is also endothermic peak of β-CD at 311°C, possibly due to thermal decomposition. The Plai oil scan demonstrated edothermic peaks at 100, 180 °C which may relate to its oxidation. The inclusion complexes showed different DSC results. The first endothermic peak shifted to lower temperatures (90-100°C) and the second endothermic peak also showed a little shifted to lower temperatures (295-300°C) compared to that of β-CD. These changes could be an indication of an embedded guest molecule in the β-CD cavities in the inclusion complex preparation [13].

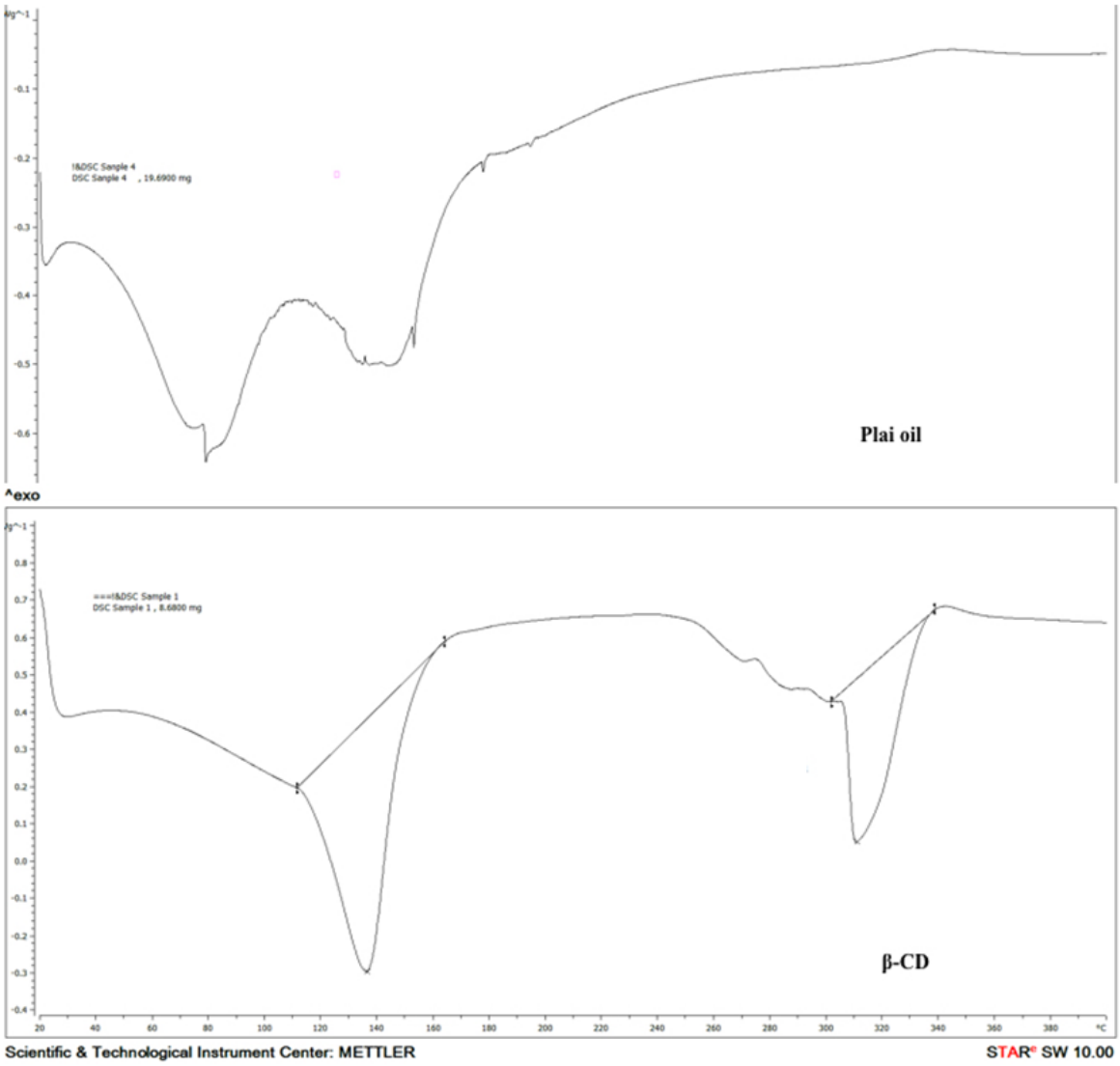

Figure 4. DSC thermograms of β-CD and Plai oil

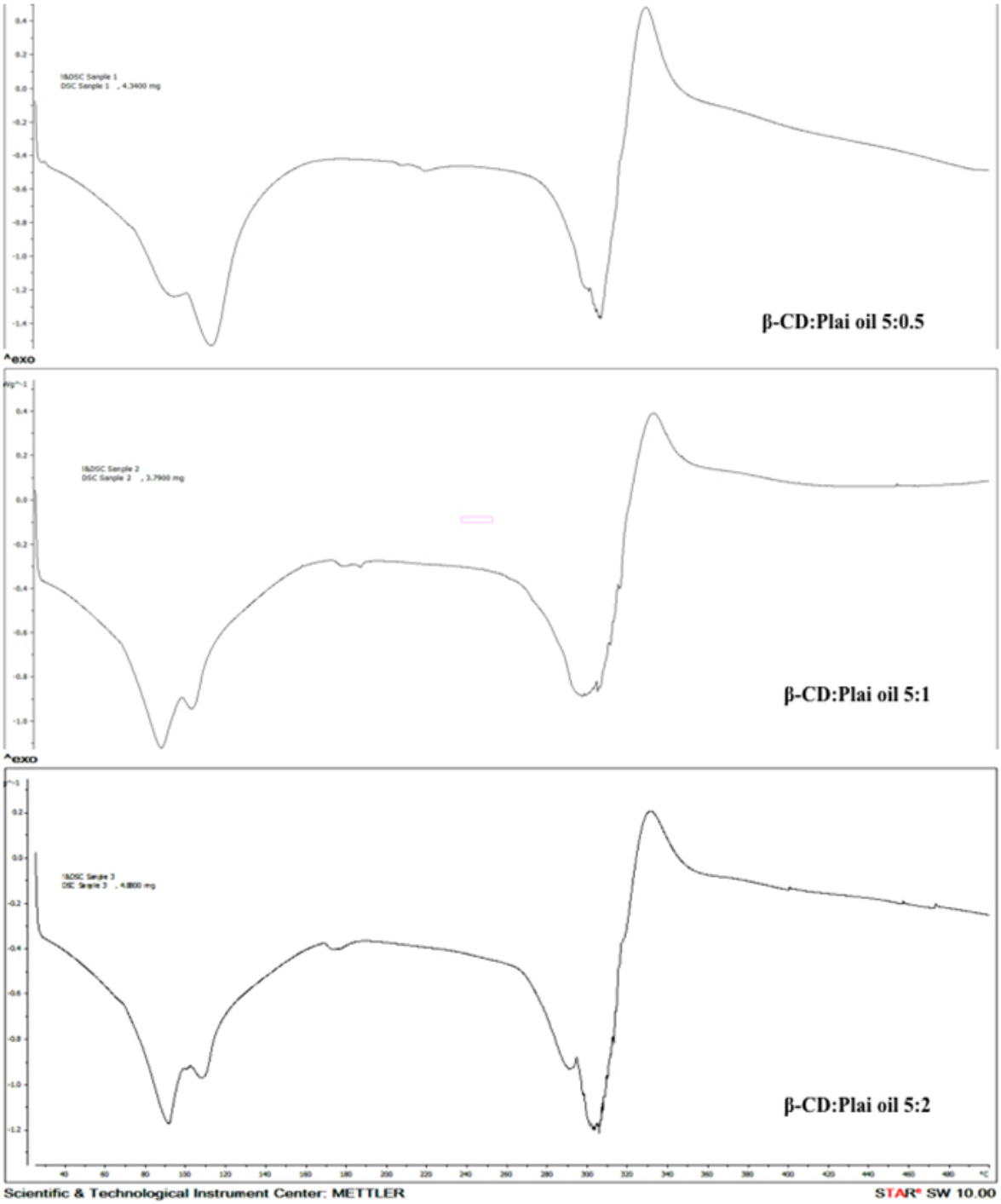

Figure 5. DSC thermograms of the Plai oil-β-CD inclusion complexes

Conclusion

The results of this study demonstrated that Plai oil could be efficiently complexed with β-CD to form an inclusion complex by a simple co-precipitation method. The results of FT-IR and DSC, demonstrated that Plai oil-β-CD complex has different physicochemical characteristics from free β-CD and Plai oil.

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