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## Original Article

# Convenient isolation of strictinin-rich tea polyphenol from Chinese green tea extract by zirconium phosphate



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## ABSTRACT

Zirconium phosphate (ZrP) was prepared and employed to separate strictinin-rich tea polyphenol from Chinese green tea extracts. The influences of ZrP calcination temperatures, green tea extraction conditions, and the amounts of ZrP on the isolation of strictinin-rich tea polyphenol were evaluated; the absorption and desorption dynamics of strictinin on ZrP were also determined. Our results revealed that the HPLC content of strictinin increased from 4.96% in 70% ethanol extract of green tea to 58.2% in isolated strictinin-rich tea polyphenol obtained by ZrP-900 (ZrP calcined at 900°C). Furthermore, the suitable time for both strictinin absorption and desorption was 4 hours at 37°C. The method developed here consisted of easy steps such as ZrP absorption, water washing, and 0.4% phosphoric acid solution desorption, which may facilitate the detection and isolation of strictinin from different samples.

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## 1. Introduction

Green tea produced from the buds of the *Camellia Sinensis* is one of the most popular beverages around the world [1]. Due to the abundance of phenolic compounds (known as tea polyphenol), green tea has plentiful health functions, including antioxidant, antiallergic, and antiviral effects [2–4]. Among these functions, tea polyphenol has received increasing attention because of its

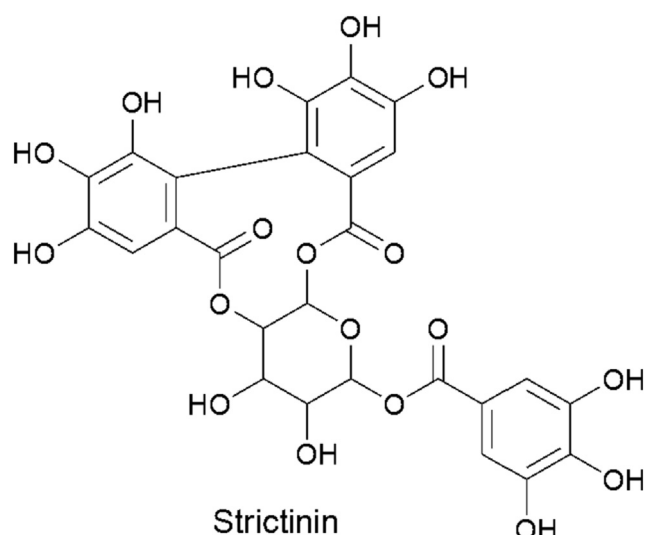
antiviral effects [5–7]. Strictinin (Figure 1), an important polyphenol found in green tea and other plants [8–10], has been proved to show special antiviral effect on influenza virus [11,12], making it a potential functional food additive. However, strictinin is one of the minor tea polyphenol in green tea [11], and the extremely low content may greatly limit its antiviral effect. Thus, increasing the content of strictinin in tea polyphenol is of great importance. Traditional methods to obtain strictinin-rich tea polyphenol include fresh silica gel

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**Figure 1** – Chemical structure of strictinin.

chromatography and high-performance liquid chromatography (HPLC) [11,13], either of which is suffered from limitations as organic solvent consuming or instrument dependent. Therefore, more facile and green method should be developed to produce strictinin-rich tea polyphenol.

Zirconium phosphate (ZrP), one of the layered materials with acidic property, has been widely applied in catalysis, ion exchange, and adsorption [14–16]. Because of its nanoscaled structure and the positive charged zirconium (IV), ZrP and their analogs (e.g., zirconium silicate) were recently proved to be special adsorbents for various phenolic compounds, including 2-chlorophenol from waste water [17], galloyl- and caffeoylquinic acids from *Galphimia glauca* and *Arnicae flos* [18], and 5-O-galloylquinic acid (a polyphenol with leishmanicidal activity) from green tea (unpublished data, Figure S1). And the latter application provides a new way to separate bioactive phenolic compounds from various plants. Similar to 5-O-galloylquinic acid, the structure of strictinin contains several galloyl groups; therefore, we assume that ZrP with special structures may be a selective adsorbent for separation of strictinin-rich tea polyphenol.

In this study, strictinin-rich tea polyphenol was conveniently isolated from green tea extracts by ZrP. The effects of material calcination temperatures, green tea extraction conditions, and material amounts on strictinin-rich tea polyphenol isolation were evaluated; the adsorption and desorption dynamics of strictinin on ZrP were also determined. A facile method for isolation of strictinin-rich tea polyphenol from green tea extract was developed.

## 2. Materials and methods

### 2.1. Materials

Green tea was bought from the local tea market (Xuancheng, China), and the standard compounds such as 5-O-galloylquinic acid, caffeine, strictinin, epigallocatechin gallate (EGCG), and epicatechin gallate (ECG) were purchased from Nanjing Spring

& Autumn Biological Engineering Co., Ltd. (Nanjing, China). Syringe filters (0.45  $\mu\text{m}$ , 13 mm) were supplied by Pall (Beijing, China), and deionized water was obtained from a water purifier system (Milli-Q, Millipore Corp., MA, USA). All chemicals and solvents were of analytical or HPLC grade.

### 2.2. Synthesis of ZrP

ZrP were prepared by direct precipitation method. In a typical synthesis, zirconium oxychloride octahydrate (1.79 g) was dissolved in 100 mL deionized water, followed by adding of concentrated phosphoric acid (85%, 0.76 mL) drop wise in 15 minutes, and the resultant solution was stirred at room temperature for another 2 hours. The white precipitate was then obtained and thoroughly washed with deionized water and ethanol by centrifugation. The precipitate was dried at 80°C in an oven for 12 hours and subsequently calcined at 500°C for 1 hour to give ZrP, which was named ZrP-500. ZrP dried or calcined under various temperatures were denoted as ZrP-n (n: temperature); they were called ZrP-80, ZrP-400, ZrP-600, ZrP-700, ZrP-800, and ZrP-900, according to the various temperatures. The structures of the materials were then characterized by X-ray diffraction (XRD; DX-2700B, Haoyuan Instrument Co., LTD, Dandong, China) or Fourier-transform infrared spectroscopy (FT-IR; Cary630, Agilent Technologies, CT, USA). XRD were determined in the  $2\theta$  range of 10° to 80° with Cu K $\alpha$  radiation; the FT-IR spectrum was recorded in the range 400–4000  $\text{cm}^{-1}$  using a potassium bromide technique.

### 2.3. Preparation of green tea extracts

The ground green tea was extracted with ethanol–water mixtures at 60°C for 1 hour in an ultrasonic cleaner (KQ50B, Kunshan Ultrasonic Instrument Co., China). In a typical extraction, ground green tea (1 g) was mixed with 70% ethanol (ethanol/water, v/v, 10 mL) and sonicated for 1 hour. The resultant solution was separated using a 0.45- $\mu\text{m}$  filter and further analyzed with HPLC or liquid chromatography-mass spectrometry (LC-MS).

### 2.4. Isolation of strictinin-rich tea polyphenol

The general procedures for isolation of strictinin-rich tea polyphenol are as follows: the mixtures of ZrP and green tea extracts were shaken for several hours on a shaker (150 rpm, 37°C); after the removal of supernatants by centrifugation, different ZrP were washed with deionized water (10 mL  $\times$  5) and subsequently mixed with the phosphoric acid solution (0.4% in water, v/v, 1 mL); the mixtures were shaken for several hours; finally, different ZrP were removed by centrifugation, and desorption solutions were obtained. In a typical separation, the ratio of ZrP and green tea extract was 1:10 (0.1 g: 1 mL), and the shaking time for each section was 24 hours. To study the adsorption/desorption dynamics of strictinin on ZrP, the supernatants from each section were extracted at different time points. For the recovery study of strictinin, the final supernatants after adsorption or desorption were collected. Each experiment was repeated three times, and the corresponding samples were analyzed by HPLC. The structure of the separated strictinin was further confirmed by nuclear magnetic resonance spectroscopy

(NMR; VNMRS 600, Agilent Technologies, CA, USA) and MS (Agilent 6460, Agilent Technologies, DE, USA).

## 2.5. HPLC and LC-MS analysis

Samples as green tea extracts, adsorption supernatants, and desorption solutions were analyzed on a reverse-phase HPLC system (Agilent 1220, Agilent Technologies, CA, USA), which was equipped with an HC-C<sub>18</sub> reverse-phase column (250 × 4.6 mm, 5 μm, Agilent) and EZChrom Elite software (Agilent). The mobile phase consisted of a phosphoric acid solution (0.4% in water, v/v, solvent A) and acetonitrile (solvent B). The samples were eluted as follows: 0–14 minutes, B linearly increased from 5% to 15%; 14–25 minutes, B maintained at 15%; 25–53 minutes, B increased from 15% to 35%; 53–60 minutes, B decreased from 35% to 5%. The flow rate was 0.7 mL/minute, and UV detection was performed at 270 nm. LC-MS analysis was performed with Agilent 1260/6460 LC/MSD system; the mobile phase consisted of aqueous formic acid (0.1% in water, solvent A) and acetonitrile (solvent B), and the mass spectra were obtained using electrospray ionization in the negative ionization modes in the range of *m/z* 100–1000, the dry gas temperature was 350°C, and the gas pressure was 50 psi; the rest conditions were identical to HPLC.

## 2.6. Data analysis

### 2.6.1. Relative content

To estimate the content changes of strictinin and other main components in the green tea extracts or desorption solutions, the relative content (%) was considered.

$$\text{Relative content (\%)} = S_C/S_L \times 100$$

where  $S_C$  is the absolute HPLC peak area of the component in green tea extracts or desorption solutions, and  $S_L$  is the largest HPLC peak area of the corresponding component in the group.

### 2.6.2. HPLC content of strictinin

To describe the HPLC content changes of strictinin in different green tea extracts or desorption solutions produced from various extraction or calcination conditions, the HPLC content of strictinin was determined.

$$\text{HPLC content (\%)} = S_S/S_T \times 100$$

where  $S_S$  is the absolute HPLC peak area of the strictinin in green tea extracts or desorption solutions, and  $S_T$  is the total HPLC peak area of all components in the corresponding solutions.

### 2.6.3. Relative adsorption or desorption efficiency

To describe the adsorption/desorption process of strictinin on ZrP, the following expressions were used.

For adsorption:

$$\text{Relative adsorption efficiency (\%)} = S_A/S_O \times 100$$

where  $S_A$  is the absolute HPLC peak area of strictinin in the green tea extracts, which were extracted at different time points during the adsorption process, and  $S_O$  is the corresponding peak area in the original green tea extract.

For desorption:

$$\text{Relative desorption efficiency (\%)} = S_B/S_D \times 100$$

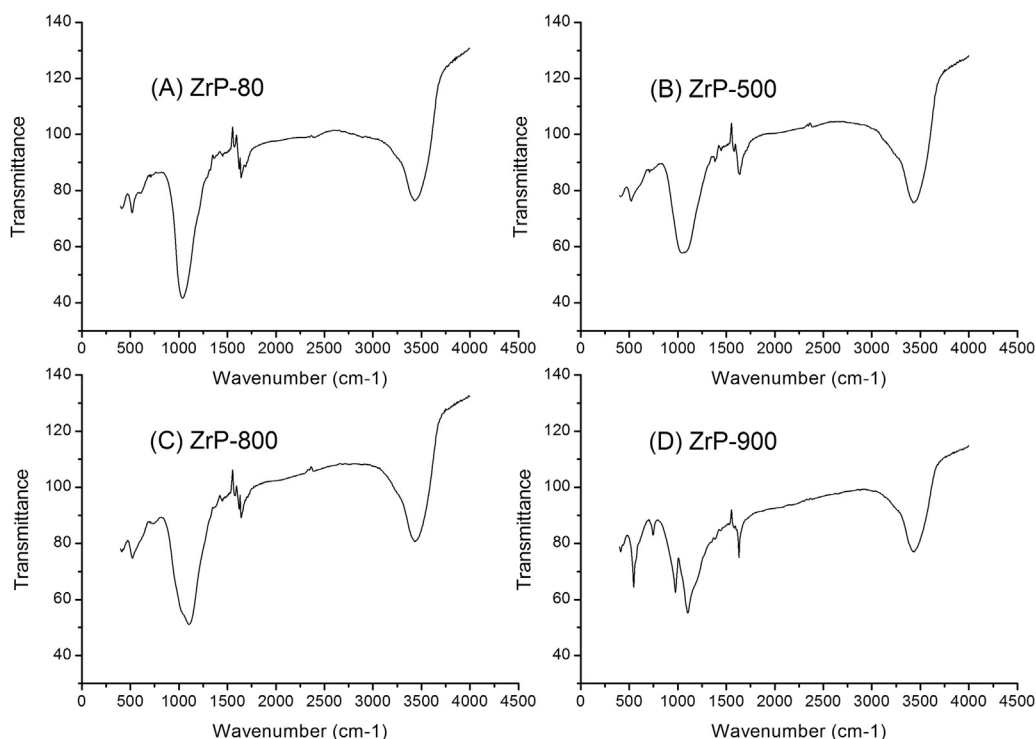
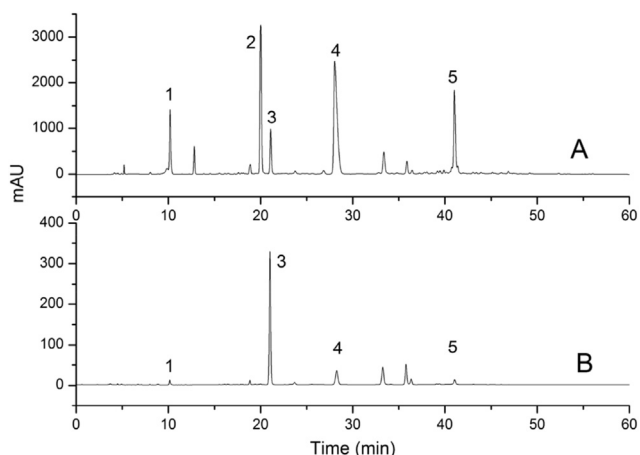


Figure 2 – FT-IR spectra of ZrP calcined under different temperatures: (A) ZrP-80, (B) ZrP-500, (C) ZrP-800, and (D) ZrP-900.



**Figure 3 – (A) HPLC profiles of 70% ethanol extract of green tea; and (B) the solution desorbed from ZrP-900. The main peaks in the profiles were identified as follows: (1) 5-O-galloylquinic acid, (2) caffeine, (3) strictinin, (4) EGCG, (5) EGG.**

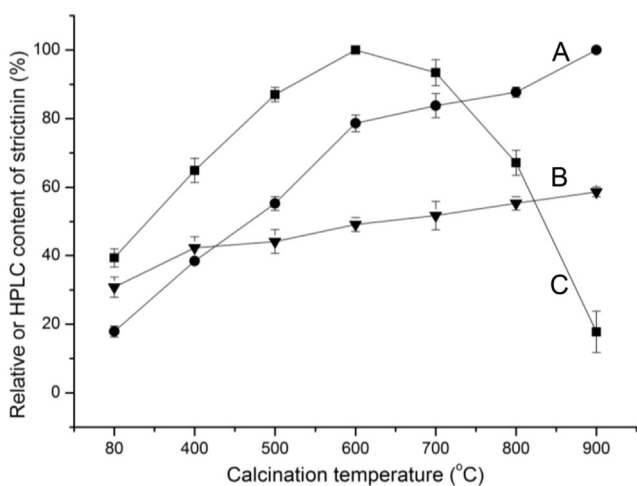
where  $S_B$  is the absolute HPLC peak area of strictinin in the desorption supernatant, which was extracted at different time points during the desorption process, and  $S_D$  is the corresponding peak area in the supernatant after 24 hours of desorption.

All of the data in Figures 3–6 are presented as the mean values of three separate experiments.

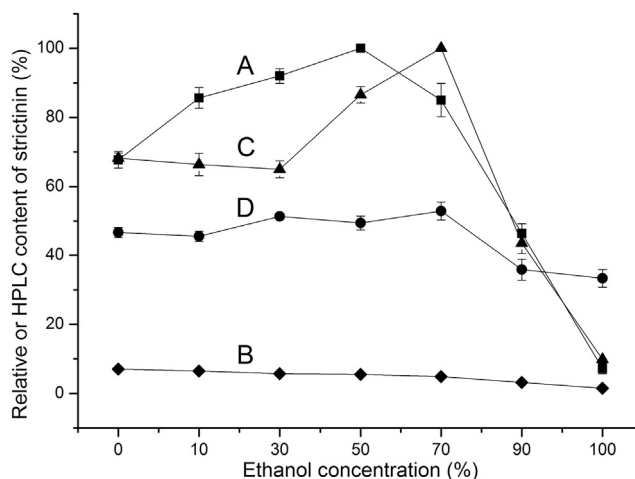
### 3. Results and discussion

#### 3.1. FT-IR characterization of ZrP

Zirconium oxychloride octahydrate and phosphoric acid were used as sources of Zr and P, respectively, for ZrP synthesis here, and the structures of the synthesized ZrP were

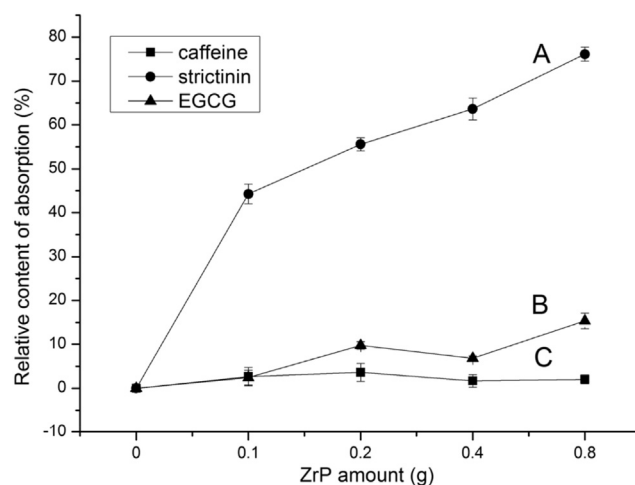


**Figure 4 – (A and B) Relative content and HPLC content of strictinin; and (C) relative content of 5-O-galloylquinic acid in solutions desorbed from ZrP that calcined under different temperatures.**



**Figure 5 – Relative content and HPLC content of strictinin in green tea extracts under different extraction conditions (A, B) and in corresponding desorption solutions (C, D).**

characterized by FT-IR. As shown in Figure 2, for ZrP dried or calcined below 900°C (ZrP-80, ZrP-500, and ZrP-800, Figure 2A–C), strong absorption bands were found at 1050  $\text{cm}^{-1}$ , 1640  $\text{cm}^{-1}$ , and 3500  $\text{cm}^{-1}$ . The strong band centered at 1050  $\text{cm}^{-1}$  due to the P–O stretching vibration; the peaks at 1640  $\text{cm}^{-1}$  and 3500  $\text{cm}^{-1}$  were attributed to OH bending vibration and asymmetric OH stretching of water molecule; the result indicated a typical structure of ZrP [19,20]. For ZrP-900 (Figure 2D), strong absorption bands were found at 540  $\text{cm}^{-1}$ , 750  $\text{cm}^{-1}$ , 980  $\text{cm}^{-1}$ , 1110  $\text{cm}^{-1}$ , 1640  $\text{cm}^{-1}$ , and 3500  $\text{cm}^{-1}$ , besides peaks at 1640  $\text{cm}^{-1}$  and 3500  $\text{cm}^{-1}$ , others were newly appeared peaks. The band at 540  $\text{cm}^{-1}$  was due to bending mode of O–P–O bond; the bands at 750  $\text{cm}^{-1}$  and 980  $\text{cm}^{-1}$  were due to symmetric and asymmetric stretching modes of P–O–P bonds, respectively, and the band at 1110  $\text{cm}^{-1}$  was due to the symmetric stretching modes of  $\text{PO}_2$  group from pyrophosphate ( $\text{P}_2\text{O}_7$ ); the results implied the appearance of  $\text{ZrP}_2\text{O}_7$  under higher calcination temperature [21,22].



**Figure 6 – Relative content of main components absorbed by different amounts of ZrP: (A) strictinin, (B) EGCG, and (C) caffeine.**

### 3.2. HPLC analysis of green tea extract and ZrP desorption solution

The content of strictinin in green tea is essential for the separation of strictinin-rich tea polyphenol. The constituent of green tea purchased from the local tea market was thus analyzed by LC-MS (Figures S2–S6). As shown in Figure 3A, the HPLC content of strictinin is 4.96%, which is higher than that of other kinds of green tea as *Longjin* and *Biluochun* (two kinds of famous green teas in China, Figure S7), making it a suitable source for strictinin-rich tea polyphenol separation. To explore the possibility of using prepared ZrP for strictinin separation, 70% ethanol extract of green tea was absorbed with ZrP-900, and the desorption solution was then analyzed by HPLC. As shown in Figure 3B, there is a main compound (peak 3) in the solution desorbed from ZrP-900, which showed a similar retention time with strictinin. However, due to the complexity of green tea components and the instability of some tea polyphenol, the main compound could not be directly identified as strictinin only on the basis of retention time. According to the MS and NMR data (Figures S3 and S8), the main compound in the desorption solution (peak 3, Figure 3B) was finally identified as strictinin. [8,23] Compared with the original extract of green tea, the HPLC content of strictinin in ZrP-900 desorbed solution achieved 57.4%, which was almost 12 times higher than its natural content as 4.96%, indicating that ZrP might be a promising absorbent for the isolation of strictinin-rich tea polyphenol

### 3.3. Effects of ZrP calcination temperatures on its strictinin adsorption properties

Compared with ZrP-900 (Figure 3B), the strictinin absorption ability of ZrP-80 was extremely low (Figure S9); thus, we supposed that the calcination temperatures of ZrP may affect their strictinin absorption properties to some degree. ZrP were then calcined under different temperatures (400–900), and their adsorption properties toward strictinin were studied. As shown in Figures 4A and 4B, with the increasing temperature of calcination, the relative content and HPLC content of strictinin in all the desorption solutions increased, and the maximum of them were obtained when using ZrP-900 as absorbent. However, an opposite result occurred with 5-O-galloylquinic acid (Figure 4C): the relative contents of that decreased with the increasing of calcination temperatures, which results may be responsible for the increased HPLC content of strictinin in desorption solutions. It is obvious that the ZrP calcination temperatures affect its strictinin adsorption properties, but the reason for that was not clear. We assumed that the changes of ZrP structure under different calcination temperatures might affect their strictinin adsorption properties, and the XRD patterns of these were then determined. As shown in Figure S10, the typical crystal structure appeared at 900°C, whereas calcination below this temperature showed no typical crystal structure, suggesting that crystal structure is essential for ZrP to absorb strictinin. The increased absorption of strictinin on ZrP during the increasing of calcination temperature (400–900) may be explained by the gradual transformation of ZrP from non-crystal structure to crystal structure [21]. To balance the

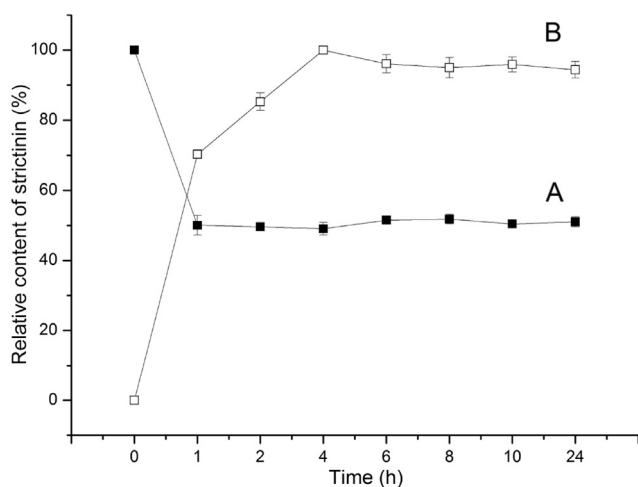
relative content and HPLC content of strictinin in desorption solution (Figure 4), ZrP-900 was chosen for further study.

### 3.4. Effects of tea extraction conditions on strictinin-rich tea polyphenol isolation

Our previous study has shown that the green tea extraction condition was essential for the absorption of 5-O-galloylquinic acid on ZrP (unpublished data), and this may be the same case for the separation of strictinin-rich tea polyphenol. Therefore, green tea extracts produced by ethanol–water mixtures containing different contents of ethanol (0–100%) were tested. As shown in Figure 5A, with the increasing concentrations of ethanol in extraction solvents, the relative content of strictinin in green tea extracts increased gradually and reached the maximum in 50% ethanol extract; however, the content declined sharply in extracts produced by solvent containing more ethanol, and the minimum content of strictinin was found in extract produced by pure ethanol, which was only 8% of the maximal one. But there are some differences for the HPLC content of strictinin under various extraction conditions (Figure 5B), which meets its maximum in pure water extract (7.6%) and its minimum in pure ethanol extract (1.6%), and the result may be due to the greater solubility of strictinin in water. To reveal the effects of different extraction conditions on strictinin-rich tea polyphenol separation, different green tea extracts were mixed with ZrP-900, and the desorption solutions produced from corresponding extracts were analyzed by HPLC. As shown in Figure 5C and D, the maximum of relative and HPLC content of strictinin (100% and 57.4%, respectively) were found in the desorption solution produced by 70% ethanol green tea extract rather than that from 50% ethanol extract (the latter tea extract contained the highest content of strictinin), which indicated that the ethanol content in extraction solvent may be an essential factor for the strictinin absorption ability of ZrP, and reason for that might be the increase or decrease of the electrostatic interaction between strictinin and ZrP in tea extracts containing different contents of ethanol [18]. Combining the relative and HPLC content together, 70% ethanol was selected as a promising extraction condition in this study.

### 3.5. Effects of ZrP amount on strictinin-rich tea polyphenol isolation

Absorbent amount is an important factor for absorption. To evaluate the effects of ZrP amount on strictinin adsorption, various amounts of ZrP (0.1–0.8 g) were mixed with 70% ethanol extract of green tea (1 mL), and the supernatants after absorption were analyzed by HPLC. As shown in Figure 6, with the increasing amount of ZrP, the relative absorption amount of strictinin and other components increased, but the increase of the latter one was not proportional to the former one. For example, when the amount of ZrP increased from 0.1g to 0.8g, the relative absorption amount of strictinin (Figure 6A) just increased from 43% to 75%, and the result may be explained by the limitation in well-mixing of the ZrP with green tea extract, indicating that liquid/solid ratio between green tea extract and ZrP is critical to strictinin adsorption. Besides, the data in Figure 6 also indicated that ZrP prepared here exhibited higher



**Figure 7 – (A) Absorption and (B) desorption dynamics of strictinin on ZrP-900.**

affinity on polyphenols (especially for strictinin) rather than non-phenolic component as caffeine (Figure 6C), because the relative amount of caffeine showed no significant increase (less than 5%) when increasing amount of ZrP was used for absorption.

### 3.6. Adsorption/desorption dynamics of strictinin on ZrP

To clarify the relationship between time and strictinin absorption/desorption process, the adsorption/desorption dynamics of strictinin on ZrP-900 were determined. As it indicated in Figure 7, the relative absorption content of strictinin increased with the time and reached the maximum after 4 hours, and prolonging the time showed no further increase in absorption, but a slight decrease. For the desorption process, the result is similar to adsorption that the maximum desorption was achieved after 4 hours and then decreased slightly with the time. The result indicated that the promising time for both absorption and desorption of strictinin on ZrP-900 was 4 hours at the temperature of 37°C.

## 4. Conclusion

Strictinin-rich tea polyphenol was conveniently obtained from green tea extract by ZrP with simple steps as ZrP absorption, water washing, and phosphoric acid solution desorption. Under the selected conditions, such as extracting green tea by 70% ethanol, calcining ZrP at 900°C, absorbing and desorbing the strictinin for 4 hours, the HPLC content of strictinin increased from 4.96% in original green tea extract to 58.2% in strictinin-rich tea polyphenol (the HPLC profile of that was almost identical to Figure 2B, data not shown), and the latter content was about 12 times higher than the former one. Due to the special affinity on strictinin, ZrP reported here may be employed as potential absorbent to detect and isolate strictinin from various plants.

## Acknowledgments

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## Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.jfda.2016.11.013>.

## Conflicts of interest

The authors declare that there are no conflicts of interest.

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