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# Methods for increasing quantity polyphenol extracts from apple lees and the development of polyphenol extracts for use as cosmetics

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We evaluated a method of extracting polyphenol from apple lees and measured the quantity of polyphenol in apple lees. We also experimentally produced cosmetics containing polyphenol extracted from apple lees to investigate the application of polyphenol extracted from apple lees as a cosmetics material.
Keywords: Apple lees, apple polyphenol, cosmetics material, method of increasing quantity

## 1. INTRODUCTION

Aomori Prefecture is Japan's leading producer of apples. This prefecture produces about 500,000 tons of apples a year. About 100,000 tons are processed as juice, with 30,000 tons of lees discharged as industrial waste after the apples are crushed into juice. Therefore, technology to produce woodceramics (a porous carbon material) by carbonizing apple lees is being investigated and developed for more advanced utilization of apple lees.

Apples also contain "polyphenol," which is a known functional material. Polyphenol is the generic term for materials with two or more hydroxyl groups linked to an aromatic hydrocarbon framework. It concerns the bitterness and astringent taste and color of plants. The components of polyphenol are classified into about 4,000 types. Polyphenol in apples has such effects as an antioxidant, antiallergenic, deodorant, in preventing tooth decay, and for whitening. Therefore, polyphenol in apple lees can be extracted for beneficial use in producing woodceramics.

In this research, we evaluated a method of extracting polyphenol from apple lees and measured the quantity of polyphenol in apple lees. We also experimentally produced cosmetics containing apple polyphenol to evaluate the application of apple polyphenol as a cosmetics material.

## 2. EXPERIMENTAL METHOD

### 2.1 POLYPHENOL CALIBRATION CURVE

#### 2.1.1. REAGENT

Phenol reagent (Kanto Chemistry Co., Inc.), sodium carbonate (Wako Pure Medicine Industry Co., Inc.), tannic acid (Wako Pure Medicine Industry Co., Inc.) apple phenon (Nikka Whiskey Co.), (+)-catechin (Sigma Chemical Co.), (-)-epicatechin (Wakou Pure Medicine Industry), acid (ICN Biomedicals, Inc.), p-coumalic acid (ICN Biomedicals, Inc.), phlorhizin (ICN Biomedicals, Inc.), and procyanidin B1 (Funakoshi)

#### 2.1.2. FOLIN-DENIS METHOD

Polyphenol reduces phosphomolybdic acid in alkali solution to produce blue color. This method uses a spectrophotometer to compare this blue color and measure the quantity of polyphenol.

First, 100 mg of tannic acid was dissolved in water to obtain 1000 ml of water solution. Then 0 ml, 2 ml, 4 ml, 6 ml, 8 ml, and 10 ml of solution were collected in 100-ml measuring flasks, respectively. Next, 10 ml of 20% sodium carbonate solution and 5 ml of a phenol reagent were added (along with water) to the solution in each measuring flask to obtain a total volume of 100 ml. The flasks were then shaken for mixture. Thirty minutes later, the absorbance for 750 nm of each sample was measured using a spectrophotometer (Shimazu UV Visibility Spectrophotometer UVmini-1240) and a cell with a

layer length of 1 cm to obtain calibration curves.

2.1.3 ASSAY METHOD

First, 10 ml of the samples listed in Section 2.2.1 were respectively collected into 100-ml measuring flasks containing 70 ml of water. Then 10 ml of 20% sodium carbonate solution and 5 ml of phenol reagent were added (along with water) to the solution in each measuring flask to obtain a total volume of 100 ml. The flasks were then shaken for mixture. Thirty minutes later, the absorbance of each solution for 750 nm was measured using the spectrophotometer. Then the quantity of polyphenol corresponding to the calibration curves obtained was determined by converting it to weight of tannic acid. The weight (g) of polyphenol in a 100-g sample was obtained with the following expression:

Polyphenol in 100g of a sample (g/100g) (converted to weight of tannic acid) =

$$\frac{\text{polyphenol (mg/100g)}}{1000} \times \text{dilution magnification}$$

2.2 HOT WATER EXTRACTION

2.2.1 SAMPLE

- Dried raw apple lees (dried at 105°C for 3 days) (Funazawa Apple Processing Center)
- Dried apple lees (apple fiber produced by Apple Fiber Nichiro)
- Roasted apple fiber (30 minutes in frying pan under medium flame)
- Heated apple fiber (at 150°C, 160°C, 170°C, 180°C, 190°C, 200°C, 210°C, 220°C, 230°C, 240°C, 250°C, 260°C, 270°C, 280°C, 290°C, 300°C, 310°C, 320°C, 330°C, 340°C, 50°C, for 30 minutes each)

2.2.2 EXTRACTION METHOD

First, 10 g of sample and 1 liter of water were placed in a flask and heated to 105°C with a mantle heater. After one hour at constant temperature, extraction was filtered through 5A-size filter paper as sample to be measured.

2.3 PRODUCTION OF COSMETIC CREAM

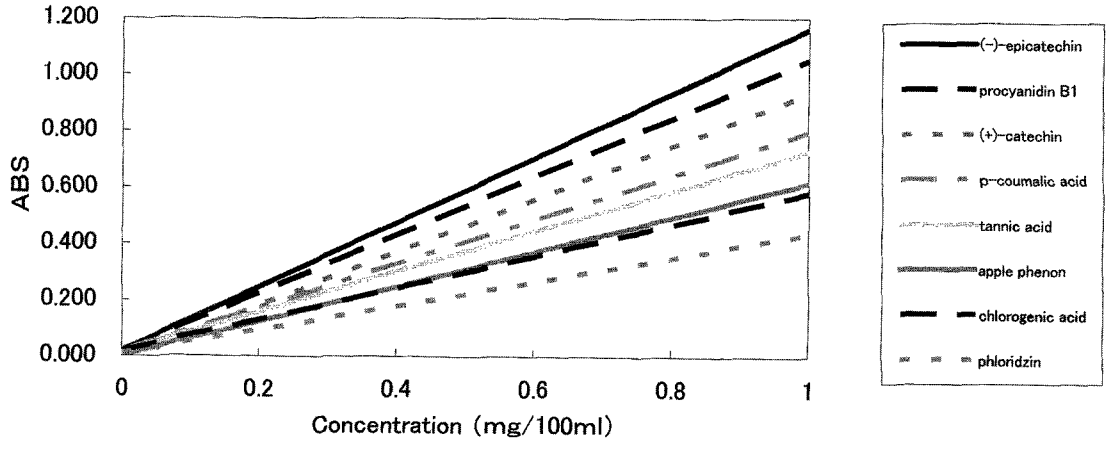
One liter of the extracted polyphenol solution was concentrated for one hour using a pressure-relief concentrator to obtain 0.17 g/100 ml of concentrated polyphenol solution. Two types of cosmetic cream containing 10% of concentrated polyphenol solution and 3% of Apple Phenon, respectively, were experimentally produced.

3 RESULTS AND DISCUSSION

3.1 POLYPHENOL CALIBRATION CURVE

Polyphenol is not a component name but a generic term. Therefore, a standard type of polyphenol must be selected to measure polyphenol. Apple contains

Fig.1 Polyphenol calibration curve



various types of polyphenol. Therefore, the quantity of apple polyphenol measured varies depending on the type of polyphenol used as standard.

Figure 1 shows the calibration curves obtained for each type of polyphenol contained in apples. As shown in this figure, different calibration curves were obtained for different types of polyphenol. Apple Phenon (a product refined from apple polyphenol) is naturally suitable as the standard. The chlorogenic acid that accounts for about 57% of apple polyphenol is also suitable because it shows a calibration curve close to that of Apple Phenon. Tannic acid is also suitable because its calibration curve represents about the average of apple polyphenol. In this research, we selected tannic acid because it is generally used in polyphenol measurement.

### 3.2 HOT WATER EXTRACTION

Figure 2 is a graph showing the quantity of polyphenol in each type of apple lee. The graph shows that about 1.0 g/100 g of polyphenol was extracted from unheated apple lees (apple fiber), and that the quantity of polyphenol in apple lees heated at and below 220° was nearly consistent. Conversely, polyphenol in apple lees heated at 230°C and 240°C increased about threefold. Its quantity in apple lees heated at 250°C showed a peak value of 3.6 g/100 g, and the quantity in apple lees heated above 250°C tended to decrease. It is conceivable that the quantity increased because the cane sugar in apple lees changed to polyphenol through caramelization. It is also conceivable that the quantity decreased because the polyphenol was gradually carbonized at over 200°C.

### 3.3. PRODUCTION OF CREAM

We experimentally produced two types of cosmetic cream containing concentrated polyphenol solution and Apple Phenon. The cream containing Apple Phenon is pink; the cream containing concentrated polyphenol solution is brown immediately after being produced. The cream containing Apple Phenon gradually became brown over time, thus showing that it is not stable. The cream containing concentrated polyphenol solution showed no significant visual change.

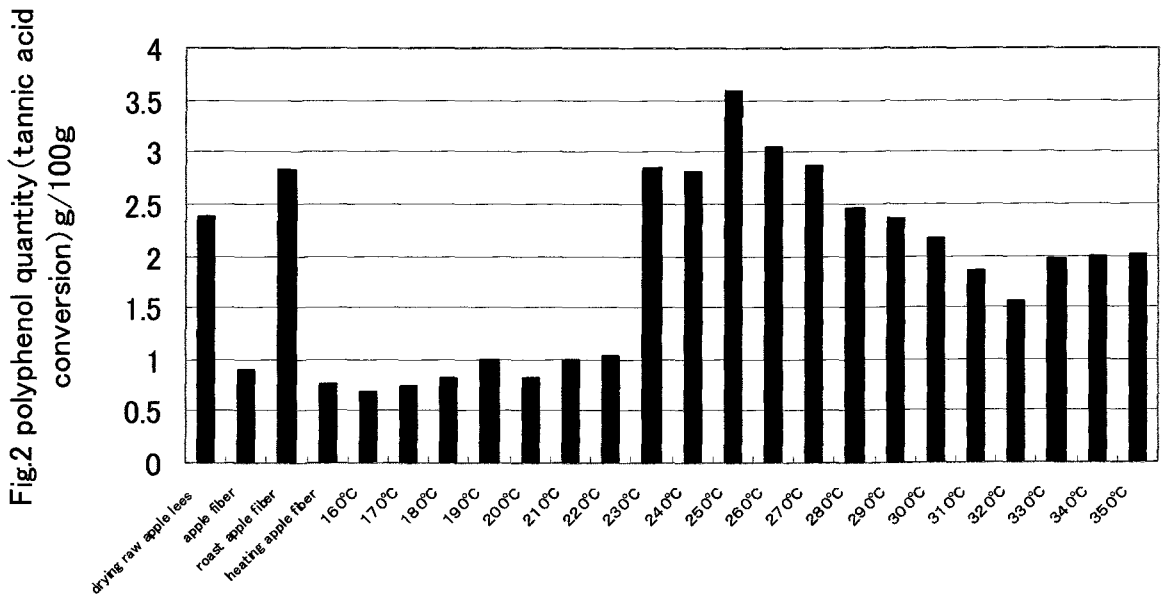
## 4. SUMMARY

### 4.1 CALIBRATION CURVE

Using the calibration curve of tannic acid, chlorogenic acid or Apple Phenon is suitable for measuring the quantity of polyphenol in apples.

### 4.2 HOT WATER EXTRACTION

Polyphenol in apple lees heated at 230°C to 250°C increases.



## 4.3 PRODUCTION OF COSMETIC CREAM

We will investigate the functionality and stability of apple polyphenol when using polyphenol as a cosmetic cream material.

REFERENCES

[1]T. Yamaguchi, Plant component analysis method, Nanedo publication, p. 287.

[2]APPLE PHENON leaflet, Nikka Whiskey Distilling Co., Inc. (development department).

[3]S. Shimoda, The characteristics and application of the apple extract (polyphenol), Food Chemical (1999-1) pp. 65-68.

[4]M. Tanabe, The characteristics and application of apple polyphenol, Food and Science (1995-1), pp. 100-103.

[5] S. Kanda, Research regarding the characteristics of the apple origin polyphenol component, positive, papers (2000).

[6]M. Yamauchi, Effects of Apple Polyphenol on Heath, Gardening Academy, northeast branch open symposia (1998).

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