Coefficient of diffusion of tannins in ethanol extracts from
Physalis alkekengi L. leaves

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Abstract. Physalis alkekengi L. (Solanaceae) is a wild growing plant, native to Bulgaria. The plant produces edible fruit rich in phytonutrients; the fruit and the leaves have been used in Bulgarian and other folk medicines for centuries. The leaves of P. alkekengi can be considered as a promising resource for obtaining extracts rich in bioactive substances [1]. The solid-liquid extraction of plant materials is performed primarily by molecular diffusion, securing the transition of the mobile bioactive plant components from the inner plant tissues to the particle surface and subsequently to the solution [7]. The mass transfer of bioactive extractibles depends on various factors involved in the process, such as the cell structure and the composition of plant tissues, the abundance and the chemical nature of the extractible substances, the polarity of the solvent, the hydro module (solid-to-liquid ratio), the temperature, duration and mode (static or dynamic) of the extraction, particle size, and others [8, 9]. A generalized indicator of the diffusive properties of the extracted plant material is the coefficient of molecular diffusion (D) [10, 11], which takes individual values for different plants and plant organs. Moreover, coefficient values change during the extraction process, due to

1. Introduction

Physalis alkekengi L. (Solanaceae) is the only species of the genus native to Bulgaria; nowadays it is found in many regions of the country as a wild growing or ornamental plant. The plant produces edible fruit rich in phytonutrients; the fruit and the leaves have been used in Bulgarian and other folk medicines for centuries [1]. In view of the current interest in natural plant products for phytopharmacy and cosmetics, the leaves of P. alkekengi can be considered as a promising resource for obtaining extracts rich in bioactive substances [2-6].
physical and chemical transformations occurring in plant tissues that alter the permeability of cell structures and other characteristics of the initial solid matrix. The huge variety of plant materials and respectively – of their characteristics and extractive behaviour, currently requires the coefficient of molecular diffusion to be determined only experimentally [12]. Calculated values of the coefficient of diffusion based on the extraction of different bioactive substances from different plant materials have been reported in a number of studies; the highly varying data between these studies results from different experimental designs and data processing procedures (e.g. technological parameters of the extraction, referenced bioactive compound(s), processed plant species and plant organs, and other variables) [9, 12-16].

To the best of our knowledge, there are no previous data about the determination of the diffusion coefficient of tannins in P. alkekengi extracts, which is set as the objective of this study.

2. Materials and methods

2.1. Plant material

Leaves from Physalis alkekengi L. plants, collected in the region of Plovdiv (Central South Bulgaria) were used in the experiments. The leaves (air-dried) were analyzed for moisture content, by drying to constant weight at 105°C, and for tannin content, by titration with potassium permanganate solution [17].

2.2. Extraction procedure and coefficients of diffusion

Extraction was carried out in a batch static mode, by maceration in the solvent, under the following conditions: raw material to solvent ratio of 1:33 (w/v); size of leaf particles – 0.0125 m × 0.002 m; duration of extraction – 1 h, with the solvent being replaced and analyzed for extracted tannins after each 600 s interval; solvents – 95%, 70%, 50%, and 30% ethanol; temperatures – 20°C, 40°C and 60°C.

The coefficient of molecular diffusion \( D \) of tannins in the ethanol extracts from P. alkekengi leaves was estimated by the following equation [18]:

\[
D = \frac{l^2 2.3 \log (E_1 - E_2)}{\pi^2 (\tau_1 - \tau_2)}
\]  

(1)

where: \( l \) – size of the material (m); \( \tau_1, \tau_2 \) – duration of extraction (s); \( E_1, E_2 \) – initial and final concentration of tannins in the solid body (%).

All experiments were carried out in threefold repetition; the mean values are presented on the figures below (created with MicroCal™ Origin 9.1 software).

3. Results and discussion

The moisture content of the air-dried leaves was 9.38±0.05%. The tannin content in the initial plant material, by exhaustive extraction, was 3.49±0.02% (on a dry weight basis).

Experimental data, plotted in Figures 1–4, revealed that the concentration of extracted tannins decreased with extraction progress; regardless of ethanol concentration and temperature, the maximum amount of tannins was extracted after the first 600-s interval. The transfer of tannins from the extracted leaves was dependent on solvent concentration and temperature, as well. In each data series, the maximum amount of tannins was extracted at 60°C, which could be attributed to the positive influence of temperature on the extractive potential of the solvent. The influence of solvent concentration upon the content of extracted tannins was relatively less pronounced, with identical maximal levels found in 30% and 50% ethanol (0.0199% and 0.0195%, 600 s, 60°C) and bigger variations, respectively, in 70% ethanol (0.0122%) and 95% ethanol (0.0287%). Those findings apparently reflected the impact of solvent nature (water-ethanol interactions with tannins), as well as that of the alterations in solid matrix structure and in the diffusion resistance within and around the particles, and other complex mechanisms on tannin extraction, related to time and temperature [19, 20].
Figure 1. Concentration of tannins (%) extracted from *P. alkekengi* leaves with 30% ethanol (temperature, 20-60°C; total duration, 1 h, 600 s intervals).

Figure 2. Concentration of tannins (%) extracted from *P. alkekengi* leaves with 50% ethanol (temperature, 20-60°C; total duration, 1 h, 600 s intervals).

Figure 3. Concentration of tannins (%) extracted from *P. alkekengi* leaves with 70% ethanol (temperature, 20-60°C; total duration, 1 h, 600 s intervals).

Figure 4. Concentration of tannins (%) extracted from *P. alkekengi* leaves with 95% ethanol (temperature, 20-60°C; total duration, 1 h, 600 s intervals).

The experimental data from each solvent extraction procedure were used for computing the diffusion coefficient values and for presenting their variation over time and temperature. The results are presented in Figures 5-8.
The highest values of the coefficient were calculated at 60°C, as follows: with 30% ethanol – 0.1607.10⁻⁹ m².s⁻¹; with 50% ethanol - 0.1612.10⁻⁹ m².s⁻¹; with 70% ethanol - 0.1690.10⁻⁹ m².s⁻¹; and with 95% ethanol - 0.1505.10⁻⁹ m².s⁻¹. As seen from those data, the numerical differences in coefficient values due to the applied solvent were minimal; still, the relatively higher values for 30%, 50% and 70% ethanol concentrations suggested better tannin diffusion compared to the 95% ethanol concentration. The coefficient of diffusion steadily decreased with time, and the highest values were recorded after the first 600-s interval; those observations were apparently related to the altered availability of the extractable substances during the process, maximal in the initial time periods and reduced by process end. Those results were in compliance with previous findings about the influence of ethanol concentration, time and temperature on the coefficient of diffusion of tannins; the obtained coefficient values, however, were lower (0.15-0.17.10⁻⁹ m².s⁻¹) than those reported in previous studies, for example with regard to P. peruviana leaves (0.28-0.31.10⁻⁹ m².s⁻¹) [16], thyme (0.51.10⁻⁹ m².s⁻¹) [13], laurel leaves (2.05.10⁻⁹ m².s⁻¹) [15] and other leaf plant materials [12, 14]. Those differences reflected the specificity of the plant material, in terms of cell structure, availability of accumulated bioactive substances in the solid matrix.

**Figure 5.** Coefficient of diffusion of tannins (D, m².s⁻¹) in extracts from *P. alkekengi* leaves obtained with 30% ethanol.

**Figure 6.** Coefficient of diffusion of tannins (D, m².s⁻¹) in extracts from *P. alkekengi* leaves obtained with 50% ethanol.

**Figure 7.** Coefficient of diffusion of tannins (D, m².s⁻¹) in extracts from *P. alkekengi* leaves obtained with 70% ethanol.

**Figure 8.** Coefficient of diffusion of tannins (D, m².s⁻¹) in extracts from *P. alkekengi* leaves obtained with 95% ethanol.
and their reproducibility into the extracts, particle size and other factors [9], as well as variations in the experimental conditions for collecting the primary data.

4. Conclusions
The study provides for the first time data about the coefficient of diffusion of tannins in ethanol extracts from Physalis alkekengi L. leaves. The highest value of the coefficient was $0.1690 \times 10^{-9}$ m$^2$.s$^{-1}$ (70% ethanol, 60°C, 600 s). The outcomes from the study contribute to the deeper insight of P. alkekengi leaf extraction and might be applied in the respective procedures for obtaining plant ethanol extracts.

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