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Improvement of extraction and stability of anthocyanins, the natural red pigment from roselle calyces using supercritical carbon dioxide extraction

Zuhaili Idham [a,b,*], Nicky Rahmana Putra [a], Ahmad Hazim Abdul Aziz [a], Ahmad Syahmi Zaini [b], Noor Azwani Mohd Rasidek [a], Norlisa Mili [b,c], Mohd Azizi Che Yunus [a,b,*]

[a] Centre of Lipids Engineering and Applied Research, Ibnu Sina Institute for Scientific and Industrial Research, Universiti Teknologi Malaysia, 81310, UTM, Johor Bahru, Malaysia
[b] School of Chemical and Energy Engineering, Faculty Engineering, Universiti Teknologi Malaysia, 81310, UTM, Johor Bahru, Malaysia
[c] Faculty of Engineering, Universiti Malaysia Sarawak (UNIMAS), Kota Samarahan, Malaysia

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ABSTRACT

In this work, anthocyanins, a natural red pigment, were isolated from roselle calyces using supercritical carbon dioxide (SC-CO_2). Three process conditions, namely pressure, temperature, and co-solvent ratio (ethanol-water), were investigated using a three-factorial design from response surface methodology (RSM). The method was used to model the extraction of anthocyanins, total phenolic content, total flavonoid content, and colour characteristics (i.e., lightness (L*), chroma (C*), and hue (h°)). The best conditions were 27MPa, 58 °C, and 8.86 % co-solvent ratio at maximal anthocyanin, phenolic, and flavonoid content, with minimal L* and C* values. The anthocyanin production was 1197 mg/100 g of dried roselle calyces. Next, the high relative value of cyanidin 3-sambubioside and phenolic compound in SC-CO_2 extract was analyzed using an ultra-high-performance liquid chromatogram (UHPLC). Anthocyanins stored at 4 °C, 25 °C, and 37 °C had average reaction rate (k) and half-life ($t_{1/2}$) values of 0.0032 and 216 days, 0.0098 and 70 days, and 0.024 and 28 days for SC-CO_2 and 0.0093 and 74 days, 0.0222 and 31 days, and 0.0444 and 15 days for solid-liquid extraction (SLE), respectively, in first-order degradation kinetics. These findings demonstrated that the studied conditions using SC-CO_2 provided lower degradation rates and a longer $t_{1/2}$ than the conventional SLE methods.

1. Introduction

Natural colourants have recently gained popularity due to the health benefits of the natural compounds used. The demand is due to several studies indicating that synthetic food colourants may have adverse effects such as high toxicity, allergic reactions, and carcinogenic potential [1]. Therefore, plant-derived anthocyanin pigments may serve as a natural substitute for synthetic red colourants. Additionally, these molecules have been linked to potential health benefits such as anti-inflammatory, antioxidant, and anti-diabetic properties [2–4]. Anthocyanins also provide excellent added value due to their dual nature as a colourant and a highly nutritious pigment.

Roselle (*Hibiscus sabdariffa*) calyces are an excellent source of anthocyanins [5]. Roselle is grown in tropical regions worldwide, including Malaysia, the Philippines, India, Mexico, and Senegal [6–8]. In Malaysia, roselle is cultivated commercially throughout the year, especially in Terengganu, Johor, Penang, Selangor, Perak, and Kedah [9]. Besides anthocyanins, roselle calyces contain many organic acids, phenolic, and glucoside compounds [3]. The recovery of anthocyanins from roselle dry calyces as a natural food colourant could expand the industry application for roselle crops.

Anthocyanins are produced in the cytoplasm and are stored in vacuoles. Several experiments have been conducted to determine the efficacy of various solvents for extracting and recovering anthocyanin compounds. Water-alcohol mixes have proven to be more efficient than pure solvent systems [10–12]. They are generally extracted using an acidified solvent extraction technique, which typically involves adding 1% acid (v/v), such as hydrochloric acid (HCl), acetic acid, or other acids to break cell walls and remove it from vacuoles [12–14]. Acids are needed to maintain a low pH environment where anthocyanins are in their more stable flavylium form [15]. This method, however, has several drawbacks, including stability issues, a long extraction time,

* Corresponding authors at: Centre of Lipids Engineering and Applied Research, Ibnu Sina Institute for Scientific and Industrial Research, Universiti Teknologi Malaysia, 81310, UTM, Johor Bahru, Malaysia.
E-mail addresses: zuhaili@cheme.utm.my (Z. Idham), r-azizi@utm.my (M.A.C. Yunus).

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