Journal of Chromatography A, 1558 (2018) 14–20

Contents lists available at ScienceDirect

Journal of Chromatography A

journal homepage: www.elsevier.com/locate/chroma

2-Naphthalenthiol derivatization followed by dispersive liquid–liquid microextraction as an efficient and sensitive method for determination of acrylamide in bread and biscuit samples using high-performance liquid chromatography

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ARTICLE INFO

Article history:
Received 15 February 2018
Received in revised form 4 May 2018
Accepted 8 May 2018
Available online 9 May 2018

Keywords:
Acrylamide
2-Naphthalenthiol
High performance liquid chromatography
Bread
Dispersive liquid-liquid microextraction
Ultrasonic-assisted extraction

ABSTRACT

In this research, an ultrasonic-assisted extraction followed by 2-naphthalenthiol derivatization and dispersive liquid-liquid microextraction of acrylamide (AA) was developed as simple and sensitive sample preparation method for AA in bread and biscuit samples using high performance liquid chromatography. Influence of derivatization and microextraction parameters were evaluated and optimized. Results showed that the derivatization of AA leads to improve its hydrophobicity and chromatographic behavior. Under optimum conditions of derivatization and microextraction, the method yielded a linear calibration curve ranging from 10 to 1000 $\mu g\,L^{-1}$ with a determination coefficient (R^2) of 0.9987. Limit of detection (LOD) and limit of quantification (LOQ) were 3.0 and 9.0 $\mu g\,L^{-1}$, respectively. Intra-day (n = 6) and inter-day (n = 3) precisions based on relative standard deviation percent (RSD%) for extraction and determination of AA at 50 and 500 $\mu g\,L^{-1}$ levels were less than 9.0%. Finally, the performance of proposed method was investigated for determination of AA in some bread and biscuit samples, and satisfactory results were obtained (relative recovery ≥ 90%).

1. Introduction

By considering of human health concern, the monitoring of contaminates in food is very important [1,2]. Acrylamide (AA) is a neo-formed contaminate (NFC), produced in food during thermal processing of raw materials used to make food by Maillard reaction of reducing sugars with asparagines at temperatures above 120 °C [3–5]. AA has been classified by the International Agency for Research on Cancer (IARC) as probably carcinogenic to humans (group 2A) [6].

Sensitive and simple analytical methods with a lower detection limits are always required for food contaminates survey. Many analytical methods for AA identification and quantification have been reported since April 2002, after declaration of its finding in food samples [7]. Due to very high polarity and solubility of AA in water, in liquid chromatography (LC) methods, its retention on C18 columns is poor. This leads to unsatisfactory separation between AA and co-extracts of matrix. However in recent years by applying hydrophilic interaction chromatography (HILIC), retention and separation of small, polar analytes on HILIC columns could be properly achieved [8]. Also, using low flow rates in analysis of AA could be leads to boarder peaks and insuffint elution of co-extracts of the matrix. Another problem in the analysis of AA in complicate matrices such as food samples is related to detection wavelength which it is generally 210 nm. At this detection wavelength, UV-absorbing compounds from co-extracts of the matrix could be interfered [9]. Also, The last problem, remain insolvable even using HILIC columns. Therefore, the most of developed methods for AA analysis have been based on GC or LC coupled with a mass spectrometer (MS or MS/MS) [10–25]. However, LC–MS/MS and GC–MS

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https://doi.org/10.1016/j.chroma.2018.05.021
0021-9673/

techniques have high sensitivity, specificity, and repeatability, but cannot satisfy the need for simple, fast and low price detection due to the requirement of expensive instruments and skilled personnel [26]. Therefore, development of sensitive methods based on GC and LC techniques are highly demand in many ordinary laboratories compared with those combined with an MS techniques [27]. Since, in recent years, derivatization with reagents such as trifluoroacetic anhydride [14,28], xanthydrol [10,11,15], and bromine [16–18] before GC and/or derivatization with reagents such as 2-mercaptobenzoic acid [19,29] and cysteine [20] before HPLC have been used for determination of AA.

Usually a separation and pereconcentration step is also required after derivatization and prior to instrumental analysis because of the trace level of AA and the complexity of foodstuff samples [10,11]. In recent years, microextraction techniques have been gaining researcher's interest [10,11,27,30–34]. Dispersive liquid-liquid microextraction (DLLME) which was introduced in 2006 by Rezaee and co-workers, is the most attractive microextraction technique [32–35]. DLLME has shown some advantages such as simplicity, high efficiency, rapidity and low cost which introduced high enrichment factor, high speed and high recovery [32].

In this research, for the first time, an ultrasound-assisted extraction followed by derivatization with 2-naphthalenthiol and dispersive liquid-liquid microextraction was developed for determining acrylamide in some food samples by HPLC-UV. Derivatization with 2-naphthalenthiol could be leads to good chromatographic of AA even on C18 column with better retention time and peak shape. At the same time, detection wavelength shifted from 210 nm to 280 nm. Both of these results lead to possibility of analysis of AA with satisfactory results by HPLC-UV consisting general C18 columns. Moreover, 2-naphthalenthiol derivatization provides possibility of microextraction of AA in oder to getting better sensitivity and clean-up. Affecting derivatization and microextraction parameters of AA were evaluated and optimized. Distinct features of the proposed method were compared with recently published researches. Finally, the proposed method was applied for determination of acrylamide in some bread and biscuit samples.

2. Material and methods

2.1. Chemicals and reagents

Analytical grade acrylamide, 2-naphthalenthiol, HPLC-grade acetonitrile (ACN), chloroform, borax, acetone, methanol, ethanol, potassium hexaferrocyanide, zinc acetate and sodium chloride were purchased from Merck Company (Darmstadt, Germany). 2-Naphthalenthiol reagent was dissolved in acetonitrile at 2.0 mg mL^{-1}. Water was purified using a Milli-Q Ultrapure water purification system (Millipore, Bedford, MA, USA). Stock standard solution of acrylamide (1000 mg L^{-1}) was prepared by dissolving proper amount of it in ultrapure water. Working standard solutions were freshly prepared by diluting of the stock solution with water. For preparation of carrez solution I, 10.6 g of potassium hexaferrocyanide was dissolved in 100 mL distillated water. Also, carrez solution II was prepared by dissolving 20 g of zinc acetate in 100 mL distillated water.

2.2. Apparatus

Chromatographic analysis was done using a high-performance liquid chromatography from Knauer of Germany model EuroChrom consisting of a degasser, quaternary pump (model K1100), manual sample injector with a 20-μL loop size, and UV detector (model K2600) was controlled by EZChrom. The injection volume was 20 μL, and column temperature was adjusted to room temperature.

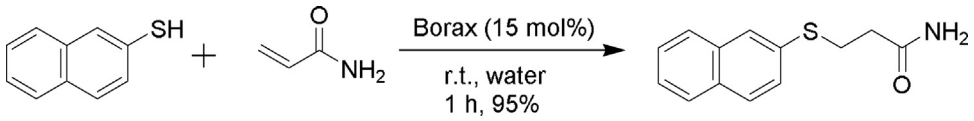

Fig. 1. Derivatization scheme of acrylamide with 2-naphthalenethiol.

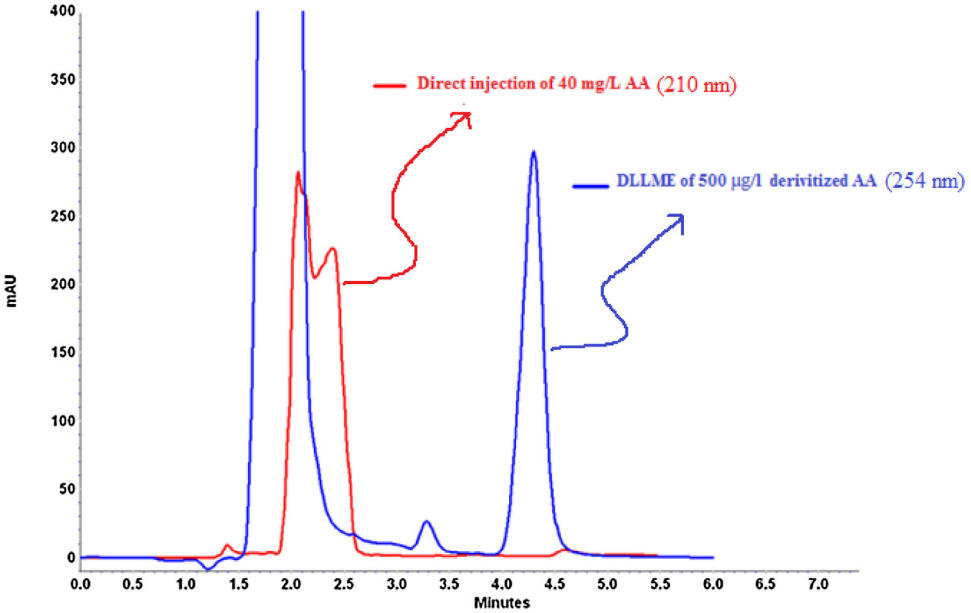

Fig. 2. Comparison of the chromatographic behavior of acrylamide before (red color chromatogram, λ = 210 nm) and after (blue color chromatogram, λ = 254 nm) derivatization and microextraction. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)