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Tandem air-agitated liquid–liquid microextraction as an efficient method for determination of acidic drugs in complicated matrices

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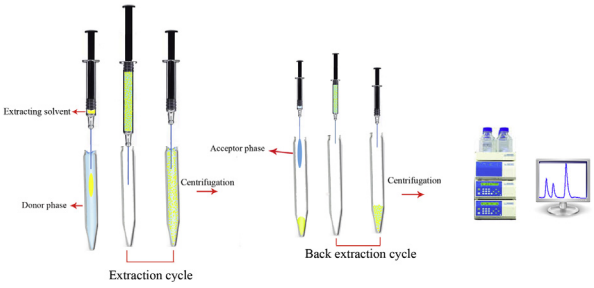
HIGHLIGHTS

- The tandem air-agitated liquid–liquid microextraction (TAALLME) method was provided.
- Some non-steroidal anti-inflammatory drugs (NSAIDs) were quantified successfully in plasma samples.
- The proposed method has the advantages of rapidity, simplicity, high efficiency, and high sample clean-up.
- The proposed method extends the application of DLLME in complicated matrices.

GRAPHICAL ABSTRACT



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ABSTRACT

A rapid and simple microextraction method with a high sample clean-up, termed as tandem air-agitated liquid–liquid microextraction (TAALLME), is described. This method is based upon the tandem implementation of the air-agitated liquid–liquid microextraction (AALLME), and this approach improves the applicability of the dispersive liquid–liquid microextraction (DLLME) methods in complicated matrices. With very simple tools, the three non-steroidal anti-inflammatory drugs diclofenac, ibuprofen, and mefenamic acid were efficiently extracted, with an overall extraction time of 7 min. By performing the first AALLME, these acidic analytes, contained in an aqueous sample solution (donor phase, 8.0 mL), were extracted into the organic solvent (1,2-dichloroethane, 37 μL), and their simple back-extraction into the aqueous acceptor solution (pH, 10.01, 51 μL) was obtained in 2 min by a second implementation of AALLME. Response surface methodology (RSM) was used for optimization of the experimental parameters. The pH values 2.94 and 10.01 were obtained for the donor and acceptor phases, respectively, and the volumes 99.5 and 51 μL were obtained for the organic solvent and the acceptor phase, respectively, as the optimal extraction conditions. Under the optimized conditions, tandem AALLME-HPLC-UV provided a good linearity in the range of 0.5–4000 ng mL^{-1}, limits of detection (0.1–0.3 ng mL^{-1}), extraction repeatabilities (relative standard deviations (RSDs) below 7.7%, n = 5), and the enrichment factors (EFs) of 80–104. Finally, the applicability of the proposed method was evaluated by the extraction and determination of the drugs under study in the wastewater and human plasma samples.

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1. Introduction

Sample pre-treatment is required prior to the analysis of most

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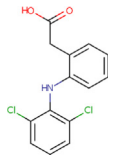
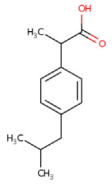
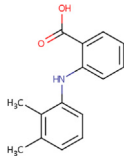
samples [1]. Hence, this step has a key position in modern analytical methodology, affecting the precision and accuracy of the final results [2]. In the past few years, the promising objectives of green chemistry have caused the conventional sample extraction procedures such as liquid–liquid extraction (LLE) and solid-phase extraction (SPE) to be overshadowed by the miniaturized and environmentally-friendly extraction methods such as liquid-phase microextraction (LPME) and solid-phase microextraction (SPME) [3]. SPME is a solvent-free extraction technique that incorporates sample concentration, sample pre-treatment, and sample introduction all into a single procedure. However, the extraction fiber is fragile and expensive, and it has a limited lifetime. Moreover, sample carry-over can be a great problem in this method [4,5]. LPME has a simple experimental set-up and a short preparation time. It is also cost-effective, and environmentally-friendly due to the consumption of low amounts of organic solvents. Single-drop microextraction (SDME) [6], hollow-fiber-protected two-phase microextraction (HF-LLME), hollow-fiber-protected three-phase microextraction (HF-LLLME) [7,8], and dispersive liquid–liquid microextraction (DLLME) are some of the most popular LPME methods [9]. Among these, HF-LLLME provides an excellent sample clean-up [10]. Despite this advantage, the method is time-consuming and an extraction may typically take 30–50 min [11]. On the other hand, DLLME provides a high enrichment and recovery in a very short period of time [12]. In this method, due to the extraction solvent being highly dispersed in the aqueous phase, the surface area between the extraction solvent and the sample solution is infinitely large, thus speeding up the extraction. Since the invention of the DLLME method in 2006 [9], continually-growing studies have been focused on this new extraction method due to its remarkable virtues, and some disperser-free methods such as the vortex-assisted liquid–liquid microextraction (VALLME) [13], ultrasound-assisted emulsification microextraction (USAEME) [14], and air-agitation liquid–liquid microextraction (AALLME) have been introduced [15,16]. Despite these considerable advantages, DLLME suffers from a major problem. A low sample clean-up is the main disadvantage of this method. The chromatograms obtained by applying this method for the preparation of samples with complex matrices, especially biological fluids, are crowded, and this intensifies distinguishing the peaks obtained for the interferences from those obtained for the analytes [17]. The distinct advantages of DLLME have caused some efforts to resolve this drawback. In this way, a growing number of studies have focused on the coalitions of DLLME with other purification or extraction techniques, exploring the compatibility with different samples, and the reports have exhibited wide combinations associating SPE [18], electromembrane microextraction (EME) [17], molecular imprinted polymer (MIP) extraction [19], stir-bar sorptive extraction (SBSE) [20], and supercritical fluid extraction (SFE) [21]. Thus a significantly higher level of selectivity was claimed to have been achieved due to the mixed approach. However, some of the mentioned methods are complicated, tedious, and time-consuming. On the other hand, the reviews carried out on the sample preparation methods have revealed that simplifying, facilitating, and reducing the consumption of organic solvents are some of the main intentions of the research efforts [12].

In the present study, a very simple, efficient, and environmentally-friendly method with a high sample clean-up was introduced for the extraction of diclofenac, ibuprofen, and mefenamic acid (the characteristics of which are shown in Table 1) from the human plasma and wastewater samples. Non-steroidal anti-inflammatory drugs (NSAIDs) (e.g. diclofenac, mefenamic acid, and ibuprofen) are commonly employed in a large scale to reduce the on-going inflammation, pain, and fever [22]. For clinical studies, it is essential to establish accurate, sensitive, and selective analytical techniques, which permit the identification and quantification of drug entities in biological samples. On the other hand, several studies carried out in different countries have shown that NSAIDs are one of the most common groups of pharmaceuticals found in effluents, and that the pharmaceutical residues are usually present in the environmental water samples in trace levels [23,24]. Hence, a sample isolation and preconcentration technique is required for the analysis of these drugs.

Table 1
Characteristics of non-steroidal anti-inflammatory drugs (NSAIDs).

Analytes	Molecular structure	Molar mass (g mol^{-1})	pK_a	Log P
Diclofenac		296.1	4.15	4.51
Ibuprofen		206.2	4.91	3.97
Mefenamic acid		241.2	4.2	5.12

For this purpose, using very simple tools, a tandem implementation of AALLME was performed, and the tandem air-agitation liquid–liquid microextraction (TAALLME) rose as a rapid method. This method is based upon a semi-three-phase extraction approach. The first AALLME provided extraction of the analytes and many interferences that could be extracted into the organic solvent. Thereby, a low sample clean-up could be obtained. In the second AALLME, the organic solvent was separated from the donor phase and exposed to an alkaline acceptor solution. Therefore, the analytes were ionized and extracted into the acceptor solution, while many interferences could not be extracted. The second step presented a simple and efficient back-extraction in less than 2 min, and in addition to the increase in the sample clean-up, the analytes were extracted into the aqueous solution. Thus this step could cause eliminating the problem of injection of the organic solvent into the final instrument analyzer as well. On the other hand, unlike most methods coupled with DLLME for improving the sample clean-up, back-extraction of the analytes was a green step in the proposed method.

2. Experimental

2.1. Chemicals and materials

Diclofenac, ibuprofen, and mefenamic acid were obtained from Sigma–Aldrich Chemie GmbH (Steinhem, Germany). Each analyte was dissolved in HPLC-grade methanol (Ameretat Shimi, Tehran, Iran) to obtain a stock solution (1 mg mL^{-1}). All the standard solutions were stored at 4 °C and re-prepared every 3 weeks. The required working standard solutions were freshly prepared by appropriate dilutions of the stock solution with deionized water to the required concentrations. The analytical-grade carbon

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