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Sodium hyaluronate-functionalized urea-formaldehyde monolithic column for hydrophilic in-tube solid-phase microextraction of melamine

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ABSTRACT

A novel sodium hyaluronate-functionalized urea-formaldehyde (UF) monolithic column has been developed by in-situ polycondensation of urea, formaldehyde and sodium hyaluronate (HA). HA plays both the roles of crosslinking and hydrophilic functionalization. The preparation factors including different molecular weights of HA and different amounts of HA were optimized, and then a uniform monolith with satisfactory permeability and hydrophilic binding capacity was obtained. Due to the excellent hydrophilicity of HA, HA-functionalized UF monolith showed higher hydrophilic extraction efficiency than UF monolith, and was applied for hydrophilic in-tube solid-phase microextraction (SPME) of melamine (MEL). Several factors for hydrophilic in-tube SPME, such as ACN percentage in the sampling solution, salt concentration and pH value of the sampling solution, elution volume, sampling and elution flow rate, were investigated with respect to the extraction efficiency of MEL. Under the optimized SPME conditions, the limit of detection (LOD) of MEL was found to be 0.2 ng/mL in the milk formula samples, the recoveries of MEL spiked in milk formula samples ranged from 87.3% to 96.7% with relative standard deviations (RSDs) less than 5.1%. Owing to the excellent hydrophilic extraction ability, the novel HA-functionalized UF monolith could provide a promising tool for the sensitive analysis of polar analytes in complicated samples.

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1. Introduction

Monolithic columns, due to its advantages of facile preparation, diverse surface functionalization, high porosity, and fast mass transfer [1–3], have attracted continuous attention in the field of analytical science. Besides the conventional application as separation stationary phase for HPLC and CEC (capillary electrochromatography), monolithic columns have also been applied as sorbents in solid-phase extraction (SPE) or solid-phase microextraction (SPME), and shown the great potential in the sample preparation for complex matrix [4–6].

Based on various hydrophilic monolithic sorbents, several hydrophilic SPME methods have been proposed and used for the efficient extraction of polar target compounds from complex samples [6–8]. Recently, a hydrophilic urea-formaldehyde (UF) monolithic column has been developed for hydrophilic in-tube SPME of polar aminoglycosides and showed a satisfactory extraction capacity towards polar anlytes [7]. Nevertheless, hydrophilic interaction chromatography column could more easily get overloaded during extraction [9], it is still necessary to further improve the extraction capacity of the monolithic sorbents for the fast and high-efficient extraction process. Since a more hydrophilic stationary phase would result in a higher hydrophilic extraction capacity [10], adding more hydrophilic substance into the polycondensation mixture should be a straightforward approach to fabricate the hydrophilic UF monolithic sorbents with higher extraction capacity. Sodium hyaluronate (HA), as a highly hydrophilic biomacromolecule, has been widely used as the drug carriers and

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the scaffolds of tissue engineering to enhance their hydrophilicity and biocompatibility [11–14]. More recently, a macroporous adsorption resin (MAR) with approximately 250 μm in diameter had been modified with HA and chitosan (CS) via a simple layer-by-layer process for improving adsorption for polar glycopeptides [15]. The resulting MAR@$(HA/CS)_{20}$ possessed highly hydrophilic property and rapid adsorption behavior. However, litter has been done on the functionalization of HA to the monolithic sorbents for hydrophilic extraction. In our previous work, a facile strategy for fabricating an ionic liquid (IL)-functionalized UF monolithic column had been proposed and turned out that the functional substance with acylamino groups could be well involved in the UF polycondensation [16]. Inspired by this work, hydrophilic HA with acylamino groups in its chemical structure should be well involved in the UF polycondensation for the hydrophilic functionalization of UF monolithic column, and further improved its hydrophilic extraction capacity.

As a highly polar analyte, melamine (MEL) is usually used to produce MEL-formaldehyde resin in plastics industry [17]. However, unscrupulous traders added MEL to milk powder or other dairy products to increase the amount of nitrogen, which will falsely show higher determinations of proteins. Infants or young children who ingested the problematic products in a high dose might induce renal failure. Thus, the development of an effective method for the routine detection of MEL was essential of substantial importance for food analysis. However, due to the low MEL concentration and the high interference of sample matrix, an effective method for sample preparation is indispensable. Among various sample pretreatment methods, in-tube SPME has been addressed the need to facilitate rapid and efficient sample pretreatment and aroused more interest [18,19]. Compared with conventional SPE, in-tube SPME possesses the advantages of solvent-free, small sample volume, simplicity, and easy automation. To date, two monolithic columns, viz. poly (2-acrylamido-2-methyl-1-propanesulfonic acid-co-ethylene dimethacrylate) (poly (AMPS-co-EDMA)) monolith [20] and attapulgite nanoparticles-modified monolith [8], have been employed for the extraction of MEL based on hydrophilic SPME. Whereas the polymerization time for these two monoliths were 12 h and 20 h, respectively. Herein, for the rapid preparation process of UF monolith, it only needs 10 min to complete the polymerization. Moreover, the report on the development of biomacromolecule functionalized monolithic column for hydrophilic in-tube SPME is still rare.

In this work, highly hydrophilic biomacromolecule HA was adopted to fabricate HA-functionalized UF monolithic column for the improvement of hydrophilic extraction capacity. So far as we aware, this is the first report on using HA for the fabrication of monolithic column. The developed monolithic columns were characterized by scanning electron microscopy (SEM), Raman spectroscopy and so on. The resulted monolithic column was then online coupled with HPLC-DAD for sensitive determination of MEL. Several factors of the online system were optimized systematically. The proposed in-tube SPME method based on HA functionalized UF monolith was successfully applied for the hydrophilic microextraction of MEL in practical milk formula samples.

2. Experimental

2.1. Chemicals and materials

Sodium hyaluronate with different molecular weights and melamine (MEL) were purchased from Sigma (St. Louis, MO, USA). Urea was supplied by Acros (New Jersey, USA). Formaldehyde solution, hydrochloric acid and toluene were obtained from Shanying Chemical Reagent (Shanghai, China). Acetonitrile (ACN) and methanol (Chemical Reagent Corporation, Shanghai, China) were of HPLC grade. Deionized water was obtained by using a Millipore Milli-Q purification system (Milford, MA, USA). Polytetrafluoroethylene (PTFE) tubes (750 μm i.d.) were obtained from Unimicro Technologies (Shanghai, China). The fused-silica capillary with dimension of 250 μm i.d. was obtained from the Refine Chromatography Ltd. (Yongnian, Hebei, China). Milk formulas were obtained from local supermarket (Fuzhou, China).

2.2. Instrumentation and analytical conditions

As shown in Fig. S1 in Electronic Supplementary Material, the in-tube SPME-HPLC system consisted of the microextraction segment, which included a Rheodyne 7725i six-port valve (valve 1), a LC-10AD pump (pump A) (Shimadzu, Kyoto, Japan) and a 0.5 mL sample loop, and the analytical segment, which included a LC-10AD pump (pump B) (Shimadzu, Kyoto, Japan), a VICI six-port valve (valve 2) with 20 cm HA-functionalized UF monolithic column and a Shimadzu SPD-M20A photodiode array detector (Shimadzu, Kyoto, Japan). The online in-tube SPME-HPLC manipulation was referred to our previous work [21] with some modifications.

In the beginning, valves 1 & 2 were initially set at LOAD positions. The sampling solution (ACN/H_2O = 50/50, v/v) was driven by pump A to flow through the monolithic column for conditioning at 0.2 mL/min. The mobile phase was driven by pump B directly through the analytical column to obtain a stable baseline for chromatographic separation. Meanwhile, the sample loop was filled with the sample solution using a syringe. When extraction began, valve 1 was directed towards INJECT position for a given time and returned to LOAD position immediately to perform extraction. The sampling solution was kept to flow through the monolithic column for 90 s in order to eliminate the residual sample solution and reduce the interference. Then, the extracted analyte was desorbed from the monolithic column by the mobile phase at a flow rate of 0.1 mL/min by simply switching the valve 2 to the INJECT position. When extraction had finished, valve 2 was switched to the LOAD position, and followed by adjusting the flow rate of the mobile phase to 0.8 mL/min for separation.

A Syncronis 5 u C18 chromatographic column (250 × 4.6 mm) from Thermo (Boston, USA) was used for the separation. Experimental conditions for the online in-tube SPME-HPLC method were optimized as followed: the mobile phase for HPLC separation was ACN/H_2O (10/90, v/v) at a flow rate of 0.8 mL/min; column temperature was 30 °C; detection wavelength was performed at 214 nm.

In addition, SEM images were obtained with NanoSEM 230 field-emission scanning electron microscope (FEI, USA). Raman spectra of the monolithic materials were carried out with inVia Reflex micro-raman spectroscopy system (Renishaw, London, England). The refrigerated centrifuge for sample preparation was purchased from Thermo Scientific (Boston, USA).

2.3. Preparation of HA-functionalized UF monolithic column

A PTFE tube was rinsed by methanol, and then dried by passing nitrogen stream. The polycondensation mixture was composed of 1 g/mL urea solution (550 mg), formaldehyde solution (450 mg), 0.2 mol/L HCl solution (100 mg) and different amount of HA with different molecular weights, which was followed our previous work with some modification [16]. After homogenization, the reaction solution was pushed into the PTFE tube. The filled tube was sealed at both ends with rubber stoppers and submerged into a thermostatic bath at 70 °C for 10 min. After that, the obtained monolithic column was rinsed using a μHPLC pump with water for 1 h, then methanol for 2 h to remove the residual materials. Finally, the pre-

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