Sensors and Actuators B 197 (2014) 254–263

Contents lists available at ScienceDirect

Sensors and Actuators B: Chemical

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

Citrate-modified silver nanoparticles as a colorimetric probe for simultaneous detection of four triptan-family drugs

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ARTICLE INFO

Article history:
Received 18 November 2013
Received in revised form 22 February 2014
Accepted 25 February 2014
Available online 6 March 2014

Keywords:
Triptan-family drugs
Citrate-capped Ag NPs
UV–visible spectrometry
DLS
TEM

ABSTRACT

We report a simple and rapid colorimetric method for selective and simultaneous detection of four triptan-family drugs (rizatriptan, naratriptan, sumatriptan, and zolmitriptan) by using citrate-capped silver nanoparticles (Ag NPs) as colorimetric sensors. The absorption spectra and solution color of citrate-capped Ag NPs undergo dramatic changes on exposure to four triptan family drugs with new surface plasmon resonance (SPR) peaks appearing at 560, 548, 520 and 570 nm for rizatriptan, naratriptan, sumatriptan, and zolmitriptan and concomitant color change from yellow to orange and to brown. This is assumed to result from the aggregation of citrate-capped Ag NPs induced by four triptan-family drugs. The UV–visible absorption spectra, FT-IR, dynamic light scattering (DLS) and transmission electron microscopic techniques were used to confirm the aggregation of citrate-capped Ag NPs-induced by four triptan-family drugs. Under the optimized conditions, good linear relationships (R^2 = 0.976–0.991) were obtained in the range of 0.001–1.0 mM, with limit of detections in the range of 7.3–84.0 nM for four triptan drugs. This method was successfully applied to detect four triptan-family drugs simultaneously in pharmaceutical samples (tablets and nasal spray) with minimized sample volumes.

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1. Introduction

Triptans are a group of tryptamine-based drugs (antimigrain agents) widely prescribed in the acute treatment of migraine headaches [1]. These drugs showed significant agonist effects on serotonin 5-HT_{1B} and 5-HT_{1D} receptors in cranial blood vessels (causing their constriction) and inhibited the release of pro-inflammatory neuropeptide in neurocells [2]. A wide range of triptan-family drugs is available in market for the treatment of migraines and cluster headaches. Among these, naratriptan, rizatriptan, sumatriptan, zolmitriptan, eletriptan, almotriptan, and frovatriptan are the most commonly used drugs for high agonist activity on the serotonin 5-HT_{1B} and 5-HT_{1D} receptors [3]. Owing to the widespread use of these drugs, a case of triptan-family drugs overdoses was carefully monitored with serial clinical observations and measurements of serum drug concentrations. The data demonstrated that the excess dose and use of triptan-family drugs cause several side effects (drowsiness, seizures, paralysis, tension of the neck, high blood pressure, legs or arms swelling, and feeling shaky) in humans [4,5]. In this connection, several analytical techniques such as capillary electrophoresis [6], voltammetry [6,7], nanocomposites electrodes-based voltammetry [8,9], high-performance liquid chromatography (HPLC) [6,10–12], liquid chromatography coupled with mass spectrometry (LC–MS) [13–15] and UV–visible spectrometric methods [6,16,17] have been used for the determination of one or more triptan-family drugs in pharmaceutical and biological samples. However, these techniques are very time consuming, and required sophisticated instrumentation, and not suitable for real-time analysis. Some methods [7,8,10,13] which can satisfy the quantitation of one triptan drug in biological fluids selectively and sensitively cannot be applied to simultaneous determination of four triptan-family drugs (rizatriptan, naratriptan, sumatriptan, and zolmitriptan). Furthermore, these techniques required ion-pair and redox complexing reagents for the analysis of triptan drugs [10–15]. Therefore, a simple, sensitive and selective method that allows simultaneous detection of four triptan-family drugs (rizatriptan, naratriptan, sumatriptan, and zolmitriptan) in pharmaceutical and in biological samples is needed.

Recently, noble metal nanoparticles-based UV–visible spectrometric methods have drawn special attention for selective and sensitive reorganization of target species (inorganic, organic and

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http://dx.doi.org/10.1016/j.snb.2014.02.087
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biomolecules) in various complex matrices [18,19]. Since, metallic NPs (Au and Ag NPs) are emerging as promising analytical colorimetric reporters for wide variety of analytes because of their intrinsically exploitable properties such as the high extinction coefficient and the distinct variation in color based on their dispersion and aggregation state [20,21]. As a result, noble metallic NPs have been used as promising coloring probes for selective, on-site and real-time colorimetric sensing of a wide variety molecules from various matrices, which facilitates to visualize targets species directly with naked eye [21]. Recent years, Ag NPs-based signal amplifications hold great promise in the development of sensitive and selective miniaturized UV–visible approaches for real-time monitoring of trace level target species in complex samples [22]. For example, Ag NPs are functionalized with various organic derivatives such as *p*-sulfonatocalix[4]arene [23–25], oligonucleotide [26], *p*-nitroaniline dithiocarbamate [27], and 4,4-bipyridine [28] and used as colorimetric sensors for detection of various organic molecules (pesticides, amino acids and DNA). These methods are based on the host–guest (electrostatic and cation–π interactions) and π–π interactions between Ag NPs and target species, yielding the color change from yellow to red or blue or green due to their induced aggregation with target analytes. To date, no attempts have made on the development of citrate-capped Ag NPs as colorimetric sensor for simultaneous detection of four triptan-family drugs in pharmaceutical samples.

Herein, we demonstrate the potential use of citrate-capped Ag NPs as colorimetric sensor for the facile, selective and simultaneous detection of four triptan-family drugs (rizatriptan, naratriptan, sumatriptan, and zolmitriptan) based on citrate-capped Ag NPs induced aggregation by four triptan-family drugs. The structures, molecular weights and solubility behavior of four-triptan family drugs are shown in Supporting Information of Table S1.

2. Materials and methods

2.1. Chemicals and materials

Silver nitrate ($AgNO_3$), sodium borohydride ($NaBH_4$), sodium citrate dihydrate, naratriptan hydrochloride (98.0%), rizatriptan benzoate (98.0%), sumatriptan succinate (98.0%) and zolmitriptan (98.0%) were purchased from Sigma–Aldrich, USA. Hydrochloric acid and methanol were purchased from Merck Ltd., India. All chemicals were of analytical grade and used without further purification. Milli-Q-purified water was used for sample preparations. Tablets (Rizatriptan benzoate, rizatriptan–5.0 mg, RIZORA 5, INTAS Pharmaceuticals Ltd., Ahmadabad, India; sumatriptan and naproxen, sumatriptan–85 mg, Lupin Ltd., Jammu, India and nasal spray, zolmitriptan–5.0 mg, Zolmist, Cipla Ltd., India) were purchased from local medical stores in Surat, Gujarat, India.

2.2. Stock solutions

Stock solutions of rizatriptan, naratriptan, sumatriptan, and zolmitriptan were prepared by dissolving 6.74, 8.34, 10.33 and 7.18 mg of each triptan drug in 25 ml of water and in 25 ml of methanol (zolmitriptan). Stock solutions were diluted with water to a final concentration drug ranging between 0.001 mM to1.0 mM.

2.3. Synthesis of citrate-capped Ag NPs

Citrate-capped Ag NPs were prepared by the reduction of $AgNO_3$ with $NaBH_4$ as a reducing agent and sodium citrate as a modifier according to the method in the literature [28,29]. Briefly, 2.0 mL of sodium citrate (50 mM) solution was added into the reaction flask that contained 78.0 mL of $AgNO_3$ (0.64 mM) solution under vigorous stirring. After 20 min, 20 mL of $NaBH_4$ (25.11 mM) was added into the above solution at room temperature and stirred for 1 h. The dark colloidal solution color was changed to bright yellow, confirming that the formation of citrate-capped Ag NPs. The citrate-capped Ag NPs solution was stored in the dark at $4.0 \pm 2.0\,^{\circ}C$ to remain stable for several weeks.

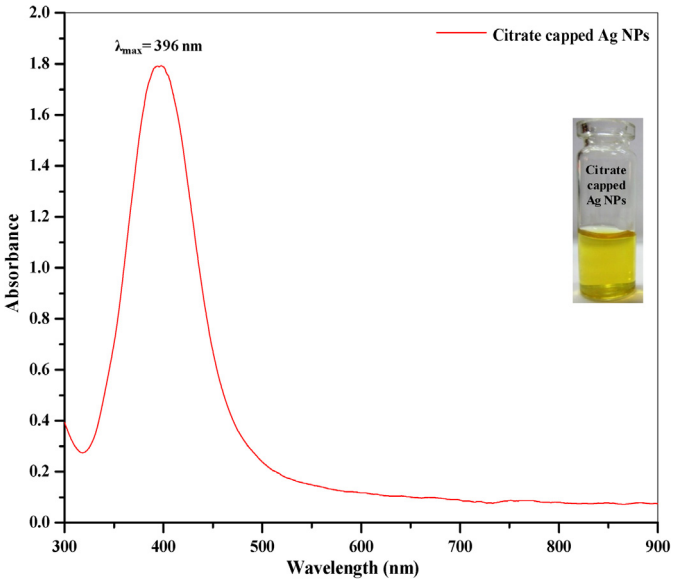

Fig. 1. UV–visible spectrum of citrate-capped Ag NPs. Inset picture show citrate-capped Ag NPs.

2.4. Citrate-capped Ag NPs as colorimetric sensor for detection of four triptan-family drugs

The citrate-capped Ag NPs-based colorimetric method was evaluated for simultaneous detection of four triptan-family drugs. Different drugs (amoxicillin, ampicilline, cortisone, cyclosporine, methylcobalamine, tramadol, naratriptan, rizatriptan, sumatriptan, and zolmitriptan; 0.7 mL, 1.0 mM) were added separately into 1.3 mL of citrate-capped Ag NPs solution at pH 8.0 and the sample vials were kept for few minutes at room temperature and the color changes were recorded by using digital camera. The UV–visible absorption spectra of the resulting solutions were recorded at different time intervals from 0 min to 3.5 min.

2.5. Instrumentation

UV–visible spectra were measured by using a Maya Pro 2000 spectrophotometer (Ocean Optics, USA) at room temperature. Fourier transform infrared (FT-IR) spectra were recorded on a PerkinElmer (FT-IR spectrum BX, Germany). Transmission electron microscopy (TEM) images were taken on a JEOL 3010. DLS measurements were performed by using Zetasizer Nano ZS90 (Malvern, UK).

3. Results and discussion

3.1. Characterization

After reduction of Ag^+ ions with sodium borohydride as a reducing agent in the presence of sodium citrate, the citrate-capped Ag NPs were characterized by UV–visible, FT-IR, TEM and DLS, respectively. Fig. 1 shows the UV–visible spectrum of citrate-capped Ag NPs. It is noticed that the characteristic SPR peak of citrate-capped Ag NPs is observed at 396 nm. Based on the UV–visible data of

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