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Design and Evaluation Sensor Starch-capped ZnSNPs Synthesis for Determination trace Tartrazine Color in Real Samples with Kinetic Spectrophotometric Method

Mostafa Ahmadi, Alireza Geramizadegan*

Department of Chemical Engineering, Dashtestan Branch, Islamic Azad University, Dashtestan, Iran

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ABSTRACT

Tartrazine is a synthetic organic food dye that can be found in common food products such as bakery products, dairy products, candies, and beverages. That the must be controlled in food produced, and content of the TZ color, potential harmful to human beings. Although the liquid chromatographic, and other methods for the TZ color has advantages such as excellent accuracy and reproducibility, it has limitations such as long-time measure, high equipment cost. In this study, for the determination TZ color in solution we used a prepared from starch-capped ZnSNPs sensor and kinetic spectrophotometric method. The calibration curve was linear in the range (0.01 to 10.0 mg L^{-1}), and the standard deviation of (1.0 %), and detection limit of the method (0.01 mg L^{-1} in time 25 min, 399 nm), were obtained for sensor level response starch-capped ZnSNPs with (95%), confidence evaluated. The chemical starch-capped ZnSNPs sensor made it possible as an excellent sensor with reproducibility.

Keywords: Color Tartrazine, Real samples, Sensor, Starch-capped ZnSNPs

1. INTRODUCTION

Wastewaters of industries like textile, paper, rubber, plastic, leather, cosmetic, food, and drug industries contain dye, and pigments which are hazardous and can cause skin irritation, cancer, and mutation in living organisms. Also, can be the cause of eye burns, and conjunctivitis in both human, and animals. Also, inhalation of them can affect respiratory tract with symptoms of rapid, or difficult breathing, while mouth ingestion can affect gastrointestinal tract with symptoms of burning sensation, nausea, vomiting, hyperhidrosis disorder, cognitive impairment disorder, micturition disorder, and methemoglobinemia – like syndromes [1]. Due to the toxic nature of some dyes, mutagenic impacts of them, and skin disorders (irritation and allergies), related to them, the potential pollution dyes, and their intermediates has drawn the attention of many researchers. In addition, microbial degradation of synthetic dyes to carcinogenic impact of benzidine and other aromatic compounds has been a matter of health concern [2].

Food dyes are often added to foodstuffs, and drinks in order to supply, intensify or

*Corresponding author: geramialireza42@gmail.com

restore their color advantages compared with natural dyes such as high stability to light, oxygen and pH, color uniformity, low microbiological contamination and relatively lower production costs. Tartrazine is a synthetic organic food dye that can be found in common food products such as bakery products, dairy products, candies, and beverages [3]. That the must be controlled in food produced, and content of the TZ color, potential harmful to human beings [4]. Therefore, is very important of the determination of the TZ color in commercial food products. Until now, different methods such as Chromatography [5,6]. Chromatographic by HPLC [7], Electro analytical [8,9], colorimetric [10], Oxidation [11], voltammetry [12], and flow injection [13], adsorption [14], and spectrophotometry [15,16]. Used methods have been reported among in determining of the TZ color. Regardless of how time-consuming these techniques, are yet, and they need advanced instrumentation, and are not fit analysis for real-time. Accordingly, developing a simple, quick, elective and delicate method like spectrophotometric measurement in determining of the TZ color was highlighted. In this method sensing of the TZ color was done with high sensitivity and excellent electivity for discerning and accurate reorganization of species (1-inorganic 2- organic and 3-biomolecules) in different intricate matrices, attention has been drawn to noble metal nanoparticles-based UV–visible spectrometric methods [17].

Due to the profitable application of metal nanoparticles, technologies, the have taken advantage of the nano-scale materials in a variety of fields from chemistry to medicine [18,19]. Recovery of nanoparticles from plant tissue is tedious, expensive and requires enzymes to destroy plant cellulose tissue. Therefore, used small molecular polymer substrates in low processing and large scale to prepare various metal nanoparticles. In recent years, the use of plant extracts for the preparation of metal nanoparticles has been proposed as an easy and suitable alternative to chemical and physical methods [20]. The forms, sizes, and structures of metallic nano materials which are extensively linked to their chemical, physical, and optical characteristics, set the ground for successful use of them in technologies. In this respect, the exceptional physical, chemical, and biological properties of ZnSNPs have been confirmed. This exceptionality arises from the size, form, composition, crystallinity, and structure of ZnSNPs in comparison with its bulk form [21]. The exclusive properties of ZnSNPs have application in the fields of the bio sensing, and nano medicine, pharmacy, and biomedical engineering varying sizes, and shapes have been utilized in a broad range of applications, and medical equipment, such as electronic devices, paints, coatings, soaps, detergents [22,23].

This study aimed to find a simple, fast and very sensitive method for identifying, and measuring of the TZ color by starch-capped ZnSNPs Sensor. Various effective factors such as (pH, TZ color concentration, starch-capped ZnSNPs concentration, time reaction, ionic strength, etc.) on the response of the method and obtaining the optimal test values and obtaining the linear range, detection and accuracy of the method presented in the measurement of the TZ color, and was checking accuracy of the method in real samples.

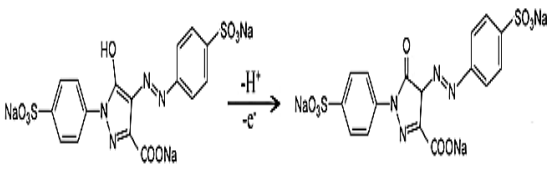

Fig. 1. Mechanism of the tartrazine (TZ) color on Starch-capped ZnSNPs.

2. EXPERIMENTAL

2.1. Reagents and materials

All chemicals of the lead nitrate $Zn(NO_3)_2$ (assay 99%), sodium sulfide (Na_2S) (assay 99.0 %), Starch, from Merck Company, and tartrazine (TZ) color (assay 98.0%), were purchased from India Company. Again, Universal buffer solutions were prepared from 1 ml of boric acid /acetic acid / phosphoric acid (1.0 M). The final pH was adjusted by the addition of 0.2M sodium hydroxide, was bought from Merck Company (Merck, Darmstadt, Germany).

2.2. Instrumentation

UV–vis spectrophotometer (Model UV–vis Shimadzu 180, Japan). The registration of FT-IR or Fourier transform infrared spectra were done on a PerkinElmer (FT-IR spectrum BX, Germany). SEM (Scanning electron microscopy: KYKY-EM 3200, Hitachi Firm, China), under an acceleration voltage of 26kV) used to study the morphology of samples. TEM images (Transmission electron microscopy), were taken on a (TEM, JEOL, Hitachi Company, China). For the measurement of pH, the pH/Ion meter (model-728, Metrohm Firm, Switzerland, Swiss), were employed.

2.3. Pretreatment of real samples

In a 50 mL beaker, treatment of a 10 mL portion of strawberry jelly (Tehran, Iran), fruity candy (Ben Ben, Iran), Smarties (morvarid, Iran), gummy candies (YupiIndonesia), noshmak (Tehran, Iran), and jell gum with fruit taste (Shiba Co, PASTIL), samples were dissolved in warm water, filtered if necessary, (or a spiked samples), and diluted in a volumetric flask. An aliquot of the above sample solutions was treated under the general procedure for the ATPS, and subsequent determination of TZ color [24].

2.4. Synthesis of Starch-capped ZnSNPs

In this regard, The nanoparticle ZnS was synthesized in reactive solution prepared using zinc nitrate $Zn(NO_3)_2$ and sulfide sodium (Na_2S) with concentration of (0.1 M and 0.1 M). The starch pellets were used as a base medium and its concentration was set to (0.1 M). 20 mL of all the above solutions were prepared separately, using distilled water as a solvent and mixed together in a beaker. The reactive vessel with solution was immersed into 20 ml acetone maintained at 40^oC and pressure of 10^{-5} mbar. A thermometer was placed in the vessel to measure the temperature of the bath solution and also a temperature sensor and dimer with temperature controller were attached to maintain the constant temperature. The solution was stirred well with the help of magnetic stirrer to maintain the homogeneous mixture. The prepared solutions, was colorless, and turned yellowish after (30 min), and suddenly changed into gray color, these indicate the chemical reactions, and also confirm the formation of the ZnS. The reactive solution was continuously stirred for (2 h). The powder was collected, and dried in a hot air oven [25].

2.6. Procedure kinetic Spectrophotometric Detection measurements

The ensuing steps have been considered for a kinetic Spectrophotometric method experiment in the current study, at the initial step: Some of the sample solution containing 1 ml of TZ color (10.0 mg L^{-1}), was added to a 10 ml volumetric balloon. Now, by increasing the first drop of 1 ml of the starch-capped ZnSNPs solution (3.0×10^{-2} $molL^{-1}$), into a balloon, the reaction start time is recorded by a timer, after 5 min from the start of the reaction the solution is stirred for 5 min, subsequently, an adequate amount of the

solution was added to a 1 cm cell. Finally, it was checked, using UV-visible spectrum (AAb), the difference between the quantities of the absorption, (399 nm), in a time interval (5.0-25.0 min).

By adding TZ color to the solution, it was observed that absorbance kinetic spectrophotometric of the starch-capped ZnSNPs at the wavelength of (399 nm) dropped. At the same time, with the help of spectrophotometry and UV–visible spectrum (AAb), the apparent spectral evolution including the formation of a well-defined isobestic point at around (399 nm) was estimated. All reaction steps were repeated by increasing the concentration (0.2 mg L^{-1}) of the TZ color every 25 min. Moreover, the mentioned steps were repeated for a reaction in the absence of TZ color (Abs b). Eventually, (Abs a) Abs blank – Abs sample was calculated. The reaction of the TZ color by starch-capped ZnSNPs was detected in the acidic medium in its wavelength (399 nm). Fig. 2, demonstrate the absorption spectra of the TZ color in solution [20,26,27].

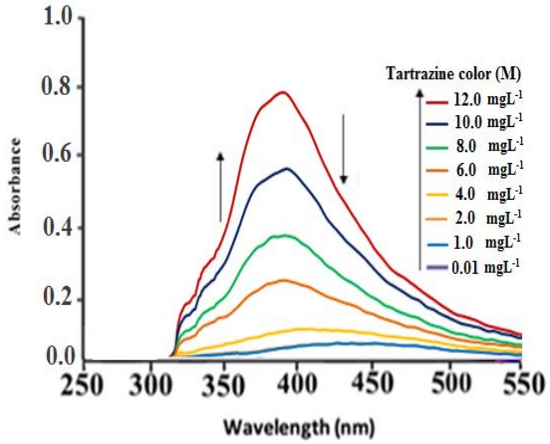

Fig. 2. The sorption spectra of result Starch-capped ZnSNPs, and TZ color 25 min, and increasing concentration of the TZ color solution (0.2 mg L^{-1}).

3. RESULTS AND DISCUSSION

3.1. Characterization of Starch-capped ZnSNPs Synthesis

FT-IR, spectra for the starch-capped ZnSNPs synthesis are shown in (Fig. 3a). The vibrational frequencies for stretching bonds in ZnS molecule cannot be detected by FTIR analysis. This confirms that ZnS doesn't show any definite absorption peaks in the range 400 - 4000 cm^{-1}. The vibration modes located at 3423 cm^{-1} can be assigned to the O–H broad absorption mode due to the hydroxyl group in the compound. The absorption band at 2928 cm^{-1} corresponds to the C-H stretching vibration mode. The broad absorption near 1300 - 1000 cm^{-1} confirms the presence of the C–O bond. The absorption band at 1637 cm^{-1} is due to the O–H bending vibration from the water molecules adsorbed into the surface. There is a furthermore subtle point that no significant difference between the FTIR spectra of the starch-capped ZnSNPs is observed [20,28]. The XRD pattern of the starch-capped ZnSNPs is shown in (Fig. 3b). The synthesized nano powders are found to be polycrystalline in nature. All detectable peaks corresponding to (111), (420), (331), (400), (222), (311), (220), (200), and (422), planes belong to the pure cubic phase of PbS (JCPDS no. 78–1901) [27]. The graph (Fig. 3c, d), is the morphological features and particle size distribution of the starch-capped ZnSNPs using SEM micrograph. It has been seen that the particles were mostly spherical with a various size distribution as they form agglomerates. From the particle size distribution, we obtain the average particle size in the range of 15-45 nm very close to those determined by XRD analysis [29].

3.2. Optimization of Sensing Conditions

In order to obtain a highly sensitive response for the detection of the TZ color the optimization of pH, buffer, starch-capped ZnSNPs, and incubation time, were carried out systematically.

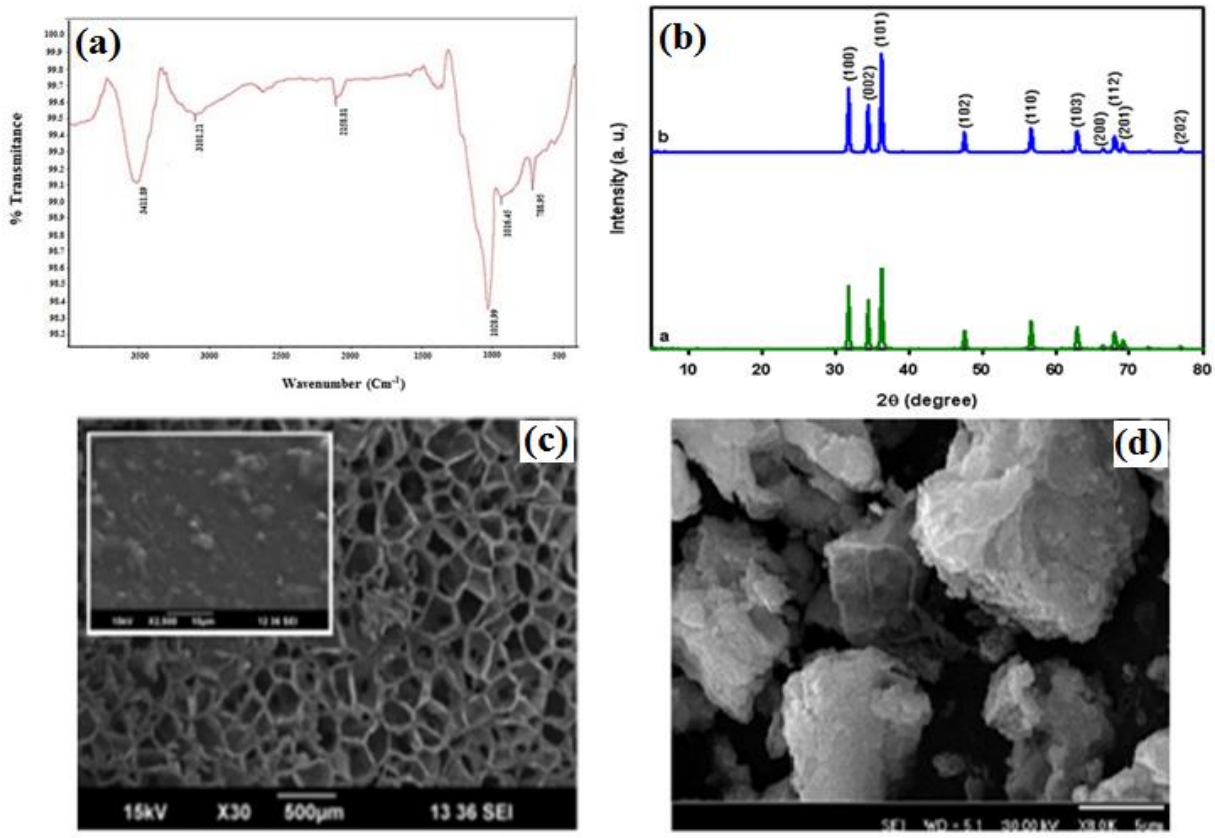

Fig. 3. (a) FT-IR spectrum image (b) XRD of the preparation of synthesized Starch-capped ZnSNPs (c) SEM image of synthesized Starch-capped ZnSNPs in 500 nm (d) SEM image of synthesized Starch-capped ZnSNPs in 100 nm.

3.2.1. Effect of buffer for rapid detection of TZ color

In this section, the best type of buffer, and its volume for maximum absorption TZ color with starch-capped ZnSNPs sensor are investigated. To this step, the procedure is as follows: In 10 ml balloons, separately 1 ml of TZ color (10.0 mg L^{-1}), and a volume of each type of acetic acid / boric acid / phosphoric acid buffer and then 1 ml of 1 ml Starch-capped ZnSNPs (2.0×10^{-2}mol L^{-1}), to the solution inside the balloon and after (25 minutes), the adsorption reaction of the solutions by the device read a spectrophotometry and UV–Visible spectrum. The results, is shown in (Fig. 4). Based on the results, 1 ml of acetic acid buffer shows the highest percentage for the determination of the TZ color, so acetic acid / tri chloric acetate buffer (1.0 M) to adjust the pH solution as the optimal buffer [28,29].

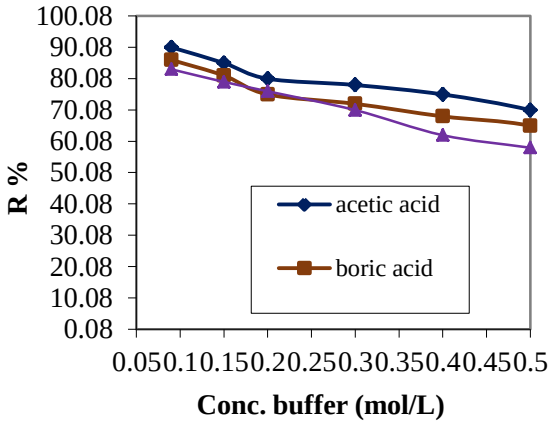

Fig. 4. The impact of buffer on the absorbance. (Sample volume, 10 mL: Starch-capped ZnSNPs, 3.0×10^{-2} M, TZ color = 10.0 mgL^{-1}, pH= 4, time 25 min, 399 nm).

3.2.2. Optimization of pH decomposition

A very important factor for measurements is the pH of the decomposition solution. To find the best pH for determining the TZ color (10.0 mg L^{-1}) with starch-capped ZnSNPs sensor (the pH range from 2 to 10, in 399 nm) were performed by method spectrophotometric kinetics for TZ color was scrutinized (Fig. 5) [28,30].

In this Study, the absorbance difference, in other words, the calculation of the difference between the absorbance of the starch-capped ZnSNPs and the absorbance of the compound TZ-Starch-capped ZnSNPs at (399 nm) would lead to absorbance measurement, absorbance increased rapidly on changing the pH from 1.0 to 4.0, while it decreased at pH values higher than 4.0. This phenomenon might be because of the weak complexion at lower pH values (pH < 4.0). On the other hand, the reduced response of the proposed starch-capped ZnSNPs sensor for the determining TZ color at pH > 4.0 could be due to a possible formation of the hydroxide of the TZ color in solution. Thus, pH 4.0 was selected as a favorable pH for all subsequent experiments [30].

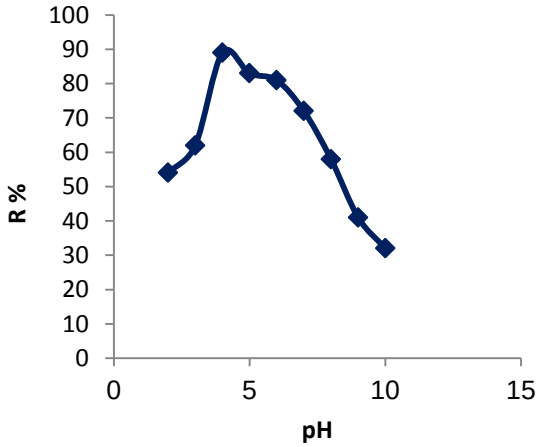

Fig. 5. The impact of pH on the absorbance. The impact of Starch-capped ZnSNPs on the absorbance. (Sample volume, 10 mL: Starch-capped ZnSNPs, 3.0×10^{-2} M, TZ color = 10.0 mgL^{-1}, time 25 min, 399 nm).

3.2.3. Impact of time on the reaction rate

For coming by the optimum time of the reaction, in a volumetric flask (10 ml) first 1 ml TZ color (10.0 mg L^{-1}) solution, 1 ml Starch-capped ZnSNPs (3.0×10^{-2}mol L^{-1}), and buffer acetic acid/ acetate (1.0 M) were added to adjust the pH solution. In the (1-10 min) interval of time, the estimation of the sorption of solutions was carried out. In other words, the calculation of the difference between the absorbance of the starch-capped ZnSNPs, and the absorbance of the compound TZ-Starch-capped ZnSNPs at (399 nm) would lead to absorbance measurement. In (Fig. 6), alterations sorption based on the time at 25^{o}C centigrade degrees temperature are exhibited. The optimum time was selected to be (25 min) [31].

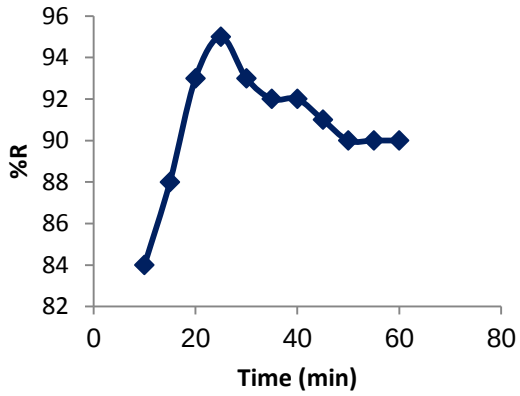

Fig. 6. The impact of contact time on the absorbance. (Sample volume, 10 mL: Starch-capped ZnSNPs, 3.0×10^{-2} M, TZ color = 10.0 mgL^{-1}, pH= 4, 399 nm).

3.2.4. Impact of Starch-capped ZnSNPs on the reaction rate

To scrutinize efficacy of the starch-capped ZnSNPs sensor on the reaction rate, 1 ml, starch-capped ZnSNPs (0.5×10^{-3} to 1.5×10^{-2} mol L^{-1}) along with 1 ml TZ color (10.0 mg L^{-1}) solution, and buffer acetic acid/ acetate (1.0 M), to adjust the pH solution, pH=4, in 399 nm), and after (25 min), the calculation of the difference

between the absorbance of the starch-capped ZnSNPs and the absorbance of the compound TZ-Starch-capped ZnSNPs at (399 nm) was scrutinized sorption of solution was estimated. As demonstrated in (Fig. 7), and by considering the results, the preferred concentration was selected to be starch-capped ZnSNPs sensor (3.0×10^{-2} mol L^{-1}) [31,32].

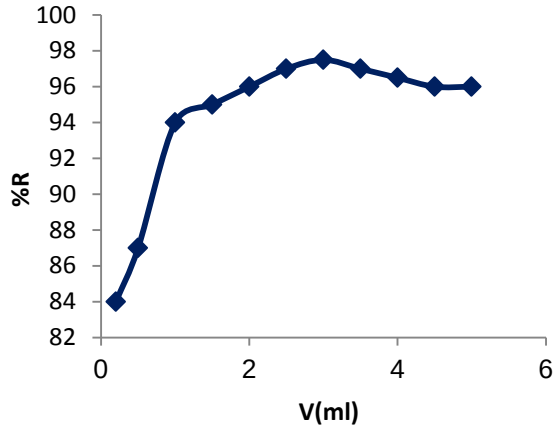

Fig. 7. The impact of Starch-capped ZnSNPs on the absorbance. (Sample volume, 10 mL: TZ color = 10.0 mgL^{-1}, pH= 4, time 25 min, 399 nm).

3.3. Analytical specifications and Calibration graph and reproducibility

After optimizing the factors affecting the measurement of TZ color, the grading curve was plotted under optimized conditions (Fig. 8). As shown the adsorption intensity in the range of TZ color (0.01-20.0 mg L^{-1}), is linearly related to the concentration of TZ color, and this error follows the equation y = 0.069x + 0.0614, where is the concentration TZ color x (mg L^{-1}), is equal to 0.9935 in terms of molar and correlation (R^2). Also, for 6 replicates, measurement of TZ color (10.0 mg L^{-1}), solution with optimized conditions, he relative standard deviation (R.S.D) for the response of the starch-capped ZnSNPs towards a (10.0 mg L^{-1}) of TZ color was (1.0 %) and reproducibility of the response of different starch-capped ZnSNPs was also studied. The determination of (10.0 mg L^{-1}) TZ color. The relative standard deviation for the response of between membranes was (1.0 %) (Fig. 8) [34,35].

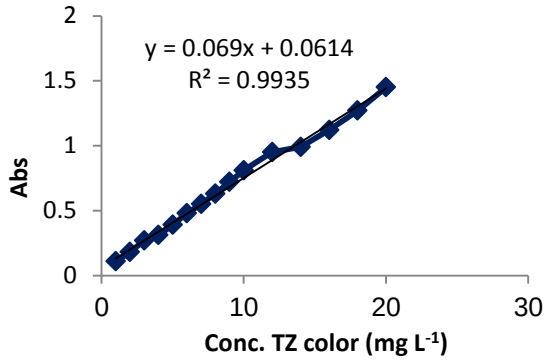

Fig. 8. The calibration graph for TZ color, on the absorbance. (Sample volume, 10 Ml, and 399 nm).

3.4. Optimum values of parameters

The optimum values of the parameters, are demonstrated in (Table. 1). The method can be used as an alternative method for the TZ color measurement owing to advantages like excellent selectivity, and sensitivity, low cost, simplicity, low detection limit and no need in utilizing organic harmful solvent.

Table 1. Investigation of method repeatability at conditions

Parameter	Optimum Value for TZ color
TZ color (M)	(10.0 mg L^{-1})
Starch-capped ZnSNPs (M)	(3.0×10^{-2} M)
pH	4.0
Equilibration time (min)	(25 min)
Linear range (LDR)	(0.01-20.0 mg L^{-1})
Detection limit (LOD)	(1.0 %)
Accuracy and precision	High
Advantages	High repeatability, sensitivity, selectivity, and wide linear range

3.5. Interference Studies

After establishing the measurement method, to evaluate the selectivity of the

prepared starch-capped ZnSNPs sensor for determining the TZ color the effect of various substances on the determination of TZ (10.0 mg L^{-1}) for method respectively was tested under optimum conditions. Tolerance Limits were defined by the concentration of interface which caused on <5% error in the determination of TZ [36]. The results showed that most of the other medications studied did not have much effect on the measurement of TZ color and among them, compounds with a more similar structure or with more functional groups are more disturbing, which It may be related to their hydrogen interactions or the molecule of the TZ color and thus reduce the measurement of the TZ color in sample. As exhibited in (Table. 2), the tolerance limit was determined as the max concentration of the interfering, substance which resulted in an error less than (±5%) for determination of the TZ color. So, selectivity of the recommended method was proven [37].

Table 2. Impacts of the matrix medicaments on the retrieving of the examined TZ color (N=6).

Foreign species	Tolerance limit (mg/L)
Sunset yellow, Glucose, Lactose, Allure red	500
Tartrate, Citrate, Oxalate, Malic acid	300
Vitamin B1, Vitamin B12, Vitamin B6	100

3.7. Application of the real sample

The complexion method was successfully applied to the determination of the TZ color in two different commercial food products (powered drink, and powdered gelatin samples) [37]. To investigate, applicability of the proposed method, recovery experiments were performed using multiple point standard addition method. This purpose, a known amount of the TZ color to formulated preparations, and the total amount of the color was estimated (n=5). The results are summarized in (Table. 3). Indicate that the prepared sensor has a very good performance for determining of the TZ color in food samples. Therefore, the determining of the TZ color in samples was confirmed utilizing standard addition method. The level of the TZ color was estimated to be below the detection limit of related element. Based on the outcomes of replicating analyses for each sample, it was shown that the medication retrievals were mainly quantitative with a low RSD. The potentiality of the recommended method for the determination of trace quantities of these elements in distinct samples was proven [38].

Table 3. Retrieval of trace TZ color in Food samples after applying presented procedure (n=5)

Samples	Added (mg L^{-1})	Founded (mg L^{-1})	RSD %	Recovery %
Smarties	0/00	0/74	1/1	----
	0/15	1/26	1/2	99
Fruity candies	0/00	0/42	1/1	----
	0/15	1/00	1/4	103
Noshmak	0/00	0/29	3/7	----
	0/15	0/80	2/8	102
Gummy candies	0/00	1/49	3/8	----
	0/15	1/97	3/0	98
Jelly stra wberry	0/00	0/44	2/3	----
	0/15	0/95	1/8	102/2
Jell Gum	0/00	0/67	2/5	----
	0/15	1/44	2/1	101/7

4. CONCLUSION

The investigation in this article focused on measuring, amount of the trace TZ color utilizing starch-capped ZnSNPs sensor. A successful analytical method for measuring TZ color was prosperously developed via utilizing a sensitized spectrophotometric with the help of starch-capped ZnSNPs. The method can be used as an alternative method for the TZ color measurement owing to advantages like excellent selectivity and sensitivity, low cost, simplicity, low detection limit, and no need in utilizing organic harmful solvent or extraction. The reaction to measuring the absorption rate of the TZ color, the optimum conditions. For determination TZ color in solution we used a prepared from

starch-capped ZnSNPs and kinetic spectrophotometric method. The calibration curve was linear in the range of (0.1 to 10.0 µg L^{-1}). The standard deviation of (1.0 %), and detection limit of the method (0.01 mg L^{-1} in time 25 min, 399 nm), were obtained for Sensor level response starch-capped ZnSNPs with (95%) confidence evaluated. The lowest determining error of the TZ color could be obtained in a short time, which strongly confirms the greater contribution for the deletion of the TZ color by starch-capped ZnSNPs sensor.

ACKNOWLEDGEMENTS

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طراحی و ارزیابی سنتز نانوذرات سولفید روی با پوشش نشاسته ای حسگر برای تعیین رنگ تارترازین در نمونه های واقعی با روش طیف سنجی جنبشی

مصطفی احمدی و علیرضا گرامی زادگان *

دانشگاه آزاد اسلامی واحد دشتستان، گروه مهندسی شیمی، واحد دشتستان، ایران

چکیده

تارترازین یک رنگ خوراکی ارگانیک مصنوعی است که در محصولات غذایی رایج مانند محصولات نانوایی، لبنیات، آب نبات‌ها و نوشیدنی‌ها یافت می‌شود، وجود و محتوای رنگ تارترازین به دلیل مضر بودن احتمالی آن برای انسان باید در محصولات غذایی کنترل شود. اگرچه کروماتوگرافی مایع، روش‌های دیگر برای رنگ تارترازین دارای مزایایی مانند دقت و تکرارپذیری عالی است، اما دارای محدودیت هایی مانند اندازه گیری طولانی مدت، هزینه بالای تجهیزات است. در این مطالعه برای تعیین رنگ تارترازین در محلول از سنسور نانوذرات سولفید روی با پوشش نشاسته‌ای و روش اسپکتروفتومتری سینتیکی استفاده شد. منحنی کالیبراسیون خطی در محدوده (۰/۰۱ تا ۱۰/۰میلی‌گرم در لیتر) بود. انحراف استاندارد (۰/۱ درصد)، و حد تشخیص روش (۰,۰۱ میلی گرم در لیتر در زمان ۲۵ دقیقه، ۳۹۹ نانومتر) برای پاسخ سطح حسگر نانوذرات سولفید روی با پوشش نشاسته با اطمینان (۹۵ درصد) ارزیابی شد. حسگر شیمیایی نانوذرات سولفید روی با پوشش نشاسته این کار را به عنوان یک حسگر عالی با قابلیت تکرار امکان پذیر کرد.

کلید واژه‌ها: رنگ تارترازین، حسگر، نانوذرات سولفید روی با پوشش نشاسته، نمونه حقیقی

* مسئول مکاتبات: geramialireza42@gmail.com