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Recognition of carbendazim fungicide in environmental samples by using 4-aminobenzenethiol functionalized silver nanoparticles as a colorimetric sensor



Gaurang M. Patel ^{a,1}, Jigneshkumar V. Rohit ^{a,1}, Rakesh Kumar Singhal ^b, Suresh Kumar Kailasa ^{a,*}

- ^a Department of Applied Chemistry, S. V. National Institute of Technology, Surat 395007, India
- ^b Analytical Chemistry Division, Bhabha Atomic Research Center, Trombay, Mumbai 400085, India

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ABSTRACT

We have developed a colorimetric method for the sensitive and selective detection of carbendazim fungicide in water and food samples using 4-aminobenzenethiol functionalized silver nanoparticles (ABT-Ag NPs) as a colorimetric sensor. The synthesized ABT-Ag NPs were characterized by UV-visible spectroscopy, FT-IR, dynamic light scattering (DLS) and transmission electron microscopic (TEM) techniques. The ABT-Ag NPs acted as a probe for binding of carbendazim through strong ion-pair and π - π interactions that causes to form a large conjugate network, resulting a color change from yellow to orange, which can be easily seen by naked eye. The characteristic surface plasmon resonance (SPR) peak of ABT-Ag NPs at 397 nm is red-shifted to 510 nm, which confirms that the carbendazim induces the aggregation of ABT-Ag NPs. Under optimum conditions, the absorbance ratio at A_{510}/A_{397} is related linearly to the concentration of carbendazim in the range of 10–100 μ M, with a detection limit of 1.04 μ M. This colorimetric method has been successfully utilized to detect carbendazim in environmental water and food samples with the recoveries ranges from 97% to 100%. The proposed colorimetric assay exhibits good reproducibility and accuracy, providing a simple and rapid method for the analysis of carbendazim in environmental water and food samples.

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1. Introduction

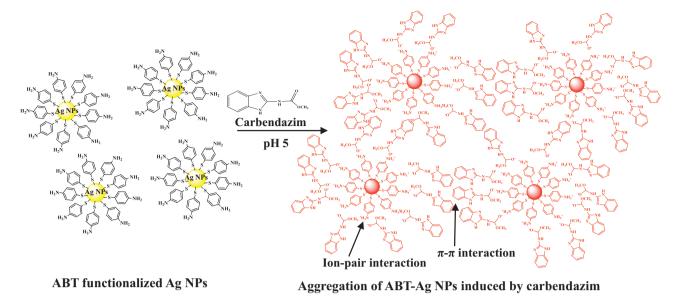
Carbendazim (methyl-1H-benzo-[d]-imidazol-2-yl-carbamate) is widely used as a benzimidazolic systemic fungicide for controlling fungal diseases in fruits crops [1]. It is also used as a casting worm controlling agent in amenity turf situations in golf greens and tennis courts [2]. Due to its high insecticidal activity, it has been used as an active ingredient in various pesticide formulations [3]. It is a major metabolite in the degradation of thiophanate and benomyl in plants [4]. Coutinho et al. have reviewed the degradation and toxic effects of carbendazim in humans and animals [5]. In this connection, it is necessary to develop a simple analytical system for the selective, sensitive and on-site detection of carbendazim in environmental samples. Several analytical methods such as UV-visible spectrometry [6], fluorimetry [7], immunoassays [8],

HPLC [9–11], mass spectrometry [12,13], voltammetry measurements [14–16] and surface enhance Raman scattering [17] have been developed for the detection of carbendazim in various environmental matrices. However, these methods require sophisticated instrumentation, complicated and time-consuming pretreatments, which limit their applications for on-site detection of carbendazim in environmental samples. Furthermore, specific buffers or ion-pairing regents are essentially needed to improve the column efficiency and reproducibility of the method. Importantly, the strong dependence of developed methods on instruments (HPLC and LC–MS) prevents them from being used in outside laboratory applications. The development of a portable platform for sensitive and selective detection of carbendazim is highly demanded.

In the era of nanoanalytical science, metallic nanoparticlesbased colorimetric sensors have gained significant interest in the development of colorimetric methods for visualization and quantification of various analytes from various samples matrices [18–21]. The integration of analytical science with nanoscience allows us to develop a promising new strategy for improving the sensitivity and selectivity of UV-visible spectrometric techniques for the identification of trace analytes with reduced sample

^{*} Corresponding author. Tel.: +91 261 2201730; fax: +91 261 2227334. *E-mail addresses*: sureshkumarchem@gmail.com, skk@ashd.svnit.ac.in (S.K. Kailasa).

¹ These authors contributed equally to this work.



Scheme 1. Schematic representation for the colorimetric detection of carbendazim using ABT-Ag NPs as a colorimetric probe.

preparations. With the emergence of nanoscience, metallic (Au and Ag) NPs have proved as potential candidates for in-situ monitoring of molecular species (inorganic, organic and biomolecules) at microvolume samples [19]. As a result, Au and Ag NPs have been used as sensors for detection of inorganic and organic pollutants with high selectivity and sensitivity [22]. For example, Cui et al. illustrated the utility of rhodamine B (RB)-capped Ag NPs as a probe for dual detection (colorimetric and fluorescence) of fenamithion with good selectivity and sensitivity [23]. Wang's and Jiang's teams developed a dual readout (colorimetric and fluorescence) method for detection of organophosphorus and carbamate pesticides using RB-capped Au NPs as a probe [24]. Li's group described the use of p-sulfonatocalix[n]arene-Ag NPs and p-sulfonatocalix[6]arene-Au NPs as colorimetric probes for sensing of pesticides and organic pollutants in water samples [25,26]. Xie et al. developed a colorimetric method for detection of organophosphates by using lipoic acid capped Au NPs as a colorimetric probe [27]. Pohanka's group developed a colorimetric dipstick method for sensing of organophosphate pesticides and nerve agents using pH papers as sensors [28]. Similarly, citratecapped Au NPs are used as colorimetric probes for detection of two organophosphorus pesticides (chlorpyrifos and malathion) and organopesticides in environmental samples with good selectivity and sensitivity [29,30]. Furthermore, Au NPs are prepared by using 3,5-dihydroxybenzoic acid as a reducer and used as a sensor for colorimetric detection of melamine in food samples [31]. Recently, Gavina et al. explored the potential application of pyridines/2-[2-(dimethylamino)phenyl]ethanol derivatives capped Au NPs as a sensor for direct colorimetric detection of organophosphorus nerve reagents and their stimulants [32]. Furthermore, Endo's group illustrated the use of polyvinylpyrrolidone coated Ag NPs as a colorimetric probe for the detection of hydrogen peroxide in environmental and food samples [33]. Rastegarzadeh and Abdali developed a novel colorimetric method for the determination of thiram pesticide by using cetyltrimethylammonium bromide capped Au NPs as a probe [34]. Moreover, our group also described that the use of 5-sulfo anthranilic acid dithiocarbamate [35] and dopamine dithiocarbamate [36] capped Ag NPs as colorimetric probes for sensing of tricyclazole and mancozeb fungicides in food and environmental samples rice samples. These reports illustrated that metallic (Au and Ag) NPs-based colorimetric approaches provide simple portable devices for on-site monitoring of trace

analytes with high selectivity and sensitivity at minimal sample volumes.

Herein, we repot the use of ABT-Ag NPs as a colorimetric probe for selective and sensitive detection of carbendazim in environmental samples. This method is based on the aggregation of ABT-Ag NPs induced by carbendazim, yielding a color change from yellow to orange (Scheme 1). In this system, ABT acts not only as a stabilizer but also as an ion-pair reagent on the surfaces of Ag NPs. As an ion-pair reagent, ABT enhances the interaction ability of Ag NPs with carbendazim, yielding ABT-Ag NPs aggregation induced by carbendazim. Additionally, the high binding force between the amino group of ABT and carbonyl (keto) group of carbendazim could keep Ag NPs stable in a complex matrix. Thus a visual sensor can be used for the sensing of carbendazim in a complex matrix, such as environmental water, fruit and vegetable samples.

2. Experimental part

2.1. Chemicals and reagents

All the reagents used were of analytical reagent grade. NaBH₄, AgNO₃, tri-sodium citrate, 4-aminobenzenethiol, sodium acetate, sodium sulphate and glacial acetic acid were purchased from Sigma–Aldrich. Technical grades of chlorpyriphos, quinalphos, indoxacarb, fipronil, clodinafop, isoprataron, sulfosulfuron and carbendazim were received from Gharda Chemicals Limited, India. Acephate and hexaconazole were received from Super Crop Safe Limited, India. Metsulfuron, imidacloprid and propiconazole were received from Atul Limited, India. Glyphosate was received from United Phosphorus Limited, India.

2.2. Synthesis of ABT-Ag NPs

The ABT-Ag NPs were synthesized by the reduction of Ag^+ ion using NaBH₄ as a reducing agent and tri-sodium citrate as a stabilizing agent. In this method, AgNO₃ (39.0 mL, 0.64 mM) and tri-sodium citrate (1.0 mL, 50 mM) solutions were added into 100 mL round-bottom flask and stirred for 20 min at room temperature. To this, freshly prepared NaBH₄ solution (10 mL, 26.43 mM) was added, and then stirred for 1 h at room temperature. The Ag NPs were functionalized with ABT by the following procedure. Briefly, 20 mL of bare Ag NPs solution was taken into 30 mL bottle that contained 0.4 mL of

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