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Naked-eye detection of potassium ions in a novel gold nanoparticle aggregation-based aptasensor

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A B S T R A C T

In this work, we studied the feasibility of interaction among gold nanoparticles (AuNPs) and a cationic dye in an aptasensor system for the detection of potassium ions. The presence and absence of potassium in the solution was distinguishable by different colors (between orange and green) appeared after reaction. Cationic dye (Y5GL) acts as a new aggregator for AuNP-based sensors which changes the aggregated AuNP solution color from blue-purple to green. In the presence of K^+ ions, the aptamer dissociated from the surface of the AuNP so that free AuNPs and cationic dye make the solution green. The aptasensor showed that the analytical linear range was from 10 nM to 50 mM and the detection limit was 4.4 nM. Also, we examined the practicality of this method on a simple paper based platform. The linear range of the colorimetric paper sensor covered of K^+ concentration from 10 μM to 40 mM and the detection limit of 6.2 μM was obtained. The selectivity of AuNP aggregation-based sensor improved by the use of cationic dye. Rapidity, simplicity, high sensitivity and excellent selectivity made this assay suitable for practical determination of K^+ in real urine samples.

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1. Introduction

Potassium is one of the most important ions needed for body homeostasis maintenance [1]. Potassium amount in the body must be regulated over a specific concentration range. Normal concentrations of potassium in the human body are between 3.5 and 5.9 mM in serum, 40–120 mM in urine, 5–10 mM in sweat and about 30 mM in saliva. Urinary potassium is for the most part secretory potassium [2]. In other words, the intake of K^+ is balanced by kidney excretion. Changes in the urinary potassium may be the result of kidney malfunction [3], neuromuscular issues, cell deaths [4], and the effect of some medications [5]. Also for those patients who have spent a long time in the intensive care unit (ICU) or those who have had severe prolonged diarrhea and vomiting or hormone problems such as hyperaldosteronism, there is a need to give them a special dosage of potassium [2].

In addition to potassium importance in biological applications, it is also a key element in soil quality determinations [6], coal combustion and flue gas cleaning systems [7] and chemical laboratories as well. In some polar drug substances, potassium determination is a part of the testing necessary for the release of the drug substance for use in clinical supplies [8]. Thus, a simple and sensitive detection of K^+ is of great significance in both clinical diagnosis and basic research.

Concentration of potassium in biological fluids is analyzed with flame photometry method [9] which not only needs special, non-portable equipment and trained operators, but also is time consuming. Other methods have been reported for potassium detection such as spectrophotometry [10], electrochemical [11], optical [12], and fluorescent [13] sensing assays. Optical sensors are extremely attractive because they minimize or eliminate the use of expensive and complicated instruments or can be easily read out by naked eye [14,15].

Recently special properties of nanomaterials have attracted enormous interest [16–18]. Although different materials such as carbon dots, Ag (silver) nanoclusters and carbon nanotubes show new features, the simplest one, gold nanoparticles (AuNPs), still have some priorities to all [19]. The synthesis process of gold nanoparticles is simple, rapid, and the product is stable almost in different conditions [20]. AuNPs are used for several purposes in medicine such as imaging, delivery of molecules into cells (i.e. siRNA, proteins and drugs) [21], diagnostic tools and biosensors. Optical biosensors coupled with AuNPs offer a powerful platform for bioanalytical sensing due to the high absorption coefficient and distance dependent color changes of AuNPs [22–24].

Aptamers are single-stranded functional oligonucleotides (DNA or RNA) which have been proved to have receptor-like activity [25]. Compared to antibodies and proteins, aptamers are chemically synthesized

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and thus more robust than antibodies, and also they can be selected against any target analyte [26–28]. Hence, combining with gold nanoparticles, a simple and accurate system can be designed for the detection of some analytes, especially ions, since the production of specific antibodies for ions is complex and costly due to the need of adjuvant [29]. Also, in comparison with potassium probe molecules; valinomycin [30] and crown ethers [31] or some probes specific for other ions [32], aptamers are easier to produce and can be obtained against all ions.

In this work, we demonstrated a new assay—using the interaction properties between gold nanoparticles and a cationic dye (Cationic Yellow 5gl)—for the highly selective and sensitive detection of potassium ions. To best of our knowledge, setting a sensitive sensor for ions is not easy because of the excess of ions already exist in any sensing solution. To increase the accuracy of the sensing platform, a cationic dye has been introduced as the AuNPs aggregator instead of salts previously reported. Despite other reported aggregation-based AuNPs systems [24], our method does not require covalent immobilization of the nucleic acid on the nanoparticle, or any addition of excess amount of salts. The color change of the solution in the presence of different K^+ concentrations is more distinguishable by the use of cationic dye. Moreover, the selectivity for K^+ ions highly improved compared to previous colorimetric sensors.

As it is known, Paper-based Analytical Devices (PADs) not only are potable, cheap and sensitive, but also can be quantified by computer software. Hence these type of sensors eliminate the need for laboratory instruments and trained operators. So we became interested to examine the feasibility of our sensing system on a PAD.

2. Experimental

2.1. Reagents and Chemicals

The potassium binding aptamer (PBA), (5′-GGG TTA GGG TTA GGG TTA GGG-3′) (100 μmol L^{-1}) used in this study was synthesized by Shanghai Sangon Biological Engineering Technology & Service Co., Ltd. Hydrogen tetrachloroaurate(III) tetrahydrate ($HAuCl_4$) (99.5%) and sodium citrate were purchased from Merck. Cationic yellow 5GL was purchased from Alibaba and other commercially available substances were purchased from Aldrich, Merck and Acros. All chemicals were of analytical grade and used without further purification. Ultra-pure water (Milli-Q plus, Millipore Inc., Bedford, MA) was used throughout the experiments.

2.2. Apparatus

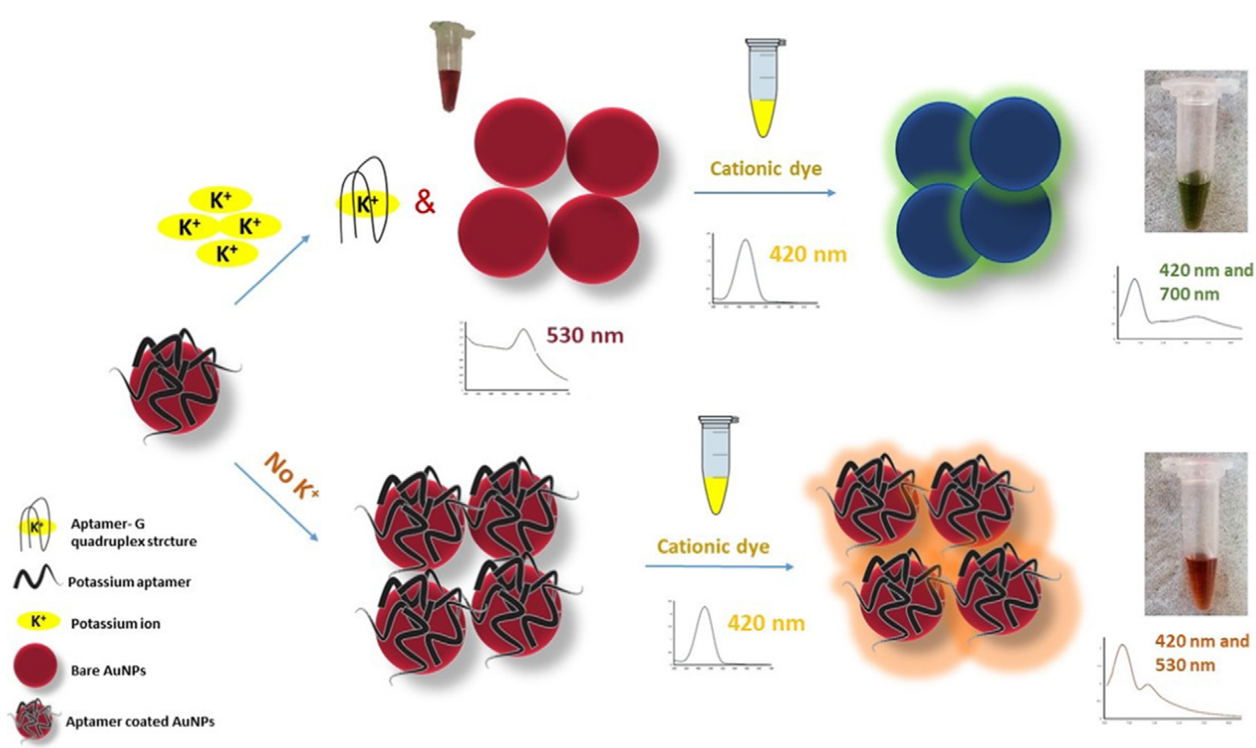
Absorption spectra were studied on a Perkin-Elmer lambda25 spectrometer. Dynamic light scattering (DLS) data were collected on a Malvern zetasizer Nano-ZS instrument.

2.3. Gold Nanoparticle Synthesis

Desired size nanoparticles prepared by thermal reduction of $HAuCl_4$ with sodium citrate as previously reported [33]. 10 mL of 39 mmol L^{-1} trisodium citrate was added to 50 mL boiling $HAuCl_4$ solution (1 mmol L^{-1}). The final solution boiled another 20 min till color changed from yellow to dark red. The characterization of synthesized AuNPs was conducted by TEM. The concentration of AuNPs was calculated using the extinction coefficient (2.7×10^8 M^{-1} cm^{-1}) at 520 nm and Beer's law to be about 10 nM. The diameter of AuNPs was about 13 nm.

2.4. Preparation of PBA-functionalized Gold Nanoparticles

To have aptamer-modified gold nanoparticles 5 μL of 2.5 μM PBA was added to 100 μL of AuNP-containing solution. Considering a recent report by Carnerero et al., the nitrogen atoms and the exocyclic amino and keto groups of DNA bases interact with AuNPs surface through physisorption and chemisorption (just for Adenosine) [34]. As the PBA is a G-rich DNA sequence, a strong interaction between AuNPs and amine bases of PBA was expected [35].



Scheme 1. Schematic illustration of the potassium sensing system.

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