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Application of the catalytic activity of gold nanoparticles for development of optical aptasensors

Rezvan Yazdian-Robati [a,1], Narges Hedayati [b,1], Shahrzad Dehghani [c], Mohammad Ramezani [d], Mona Alibolandi [d], Majid Saeedi [e], Khalil Abnous [d,f,**], Seyed Mohammad Taghdisi [g,*]

[a] Molecular and Cell Biology Research Center, Faculty of Medicine, Mazandaran University of Medical Sciences, Sari, Iran
[b] Department of Pharmacodynamics and Toxicology, School of Pharmacy, Mashhad University of Medical Sciences, Mashhad, Iran
[c] Department of Pharmaceutical Biotechnology, School of Pharmacy, Mashhad University of Medical Sciences, Mashhad, Iran
[d] Pharmaceutical Research Center, Pharmaceutical Technology Institute, Mashhad University of Medical Sciences, Mashhad, Iran
[e] Department of Pharmaceutics, Faculty of Pharmacy, Mazandaran University of Medical Sciences, Sari, Iran
[f] Department of Medicinal Chemistry, School of Pharmacy, Mashhad University of Medical Sciences, Mashhad, Iran
[g] Targeted Drug Delivery Research Center, Pharmaceutical Technology Institute, Mashhad University of Medical Sciences, Mashhad, Iran

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ABSTRACT

Biosensor technology is considered to be a great alternative in analytical techniques over the conventional methods. Among many recently developed techniques and devices, aptasensors are interesting because of their high specificity, selectivity and sensitivity. Combining aptamer as a biological recognition element with gold nanoparticles (AuNPs) as probe, are becoming more general owing to their beneficial properties, including low cost and ability to analyze specific targets on-site and using naked eye. Hydrogen bonds, nucleic acid hybridization, aptamer-target and antigen-antibody binding, Raman signature, enzyme inhibition, and enzyme-mimicking activity are main different sensing strategies exploited in AuNPs-based optical aptasensors. In this review article, we discussed the recent advances in optical aptasensors with a special emphasis on the catalytic activity property of AuNPs.

1. Introduction

Gold as an inert metal has been used technologically since ancient Roman times, where it was exploited to stain glasses a red or mauve color. However, in the 1850s, its use for biological applications was reported for the first time by Michael Faraday, who observed that optical properties of colloidal gold solutions differ from bulk gold [1]. In the last few years, gold nanoparticles (AuNPs) have attracted tremendous interest at a nanoscale size over other metallic nanoparticles. This is due to the highly favorable properties of AuNPs, such as their large surface area, shape and size-dependent optical and electronic characteristics [2]. They also possess other beneficial properties, including strong surface plasmon resonance (SPR) features, easy surface modification, a high X-ray absorption coefficient as well as their ease of synthetic manipulation [3–8]. Even though gold is usually recognized chemically inert, for the first time, in the 1980s, Haruta et al. showed that AuNPs are active catalysts when their particle size decreased under 5 nm. They used AuNPs in catalysis for the oxidation of CO to CO2 [9]. The use of colloidal AuNP solutions in catalysis has now been extended to numerous reactions including the oxidation of alcohols, toluene, cyclohexane and aldehydes, hydrogenation of aldehydes, unsaturated carbonyls and reduction of nitro groups, as well as the epoxidation of propylene, miscellaneous reactions and carbon–carbon bond formation [10–14]. Over the past ten years, the inherent catalytic properties of colloidal AuNPs have been of great interest in the area of biosensors especially optical aptasensors with the aim of improving the detection sensitivity, selectivity, and stability [14–18]. Indeed, AuNPs less than 3–5 nm in diameter have unique catalytic properties, so they can potentially mimic the enzymatic function of catalase, silicatein, esterase, peroxidase, glucose oxidase, nuclease and superoxide dismutase [19, 20]. These enzyme-like activities of AuNPs are associated with the size, morphology and surface state and functional groups of the nanoparticles

* Corresponding author.
** Corresponding author. Pharmaceutical Research Center, Pharmaceutical Technology Institute, Mashhad University of Medical Sciences, Mashhad, Iran.
E-mail addresses: abnouskh@mums.ac.ir (K. Abnous), taghdisihm@mums.ac.ir (S.M. Taghdisi).
[1] These authors contributed equally to the work.

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[21].

Usually the catalytic activity of AuNPs begins to increase as the size drops below ~3 nm. The explanation for origin of these phenomena has to be sought in different mechanisms, including quantum limitation impacts, charging of the nanomaterials, active engagement by the support either by giving oxygen to the cluster, or by the catalytic site being localized at the interface and the plenty of low-coordinated gold atoms on the smallest groups [22,23]. Several review papers have focused exclusively on the general physical features of AuNPs [24,25]. AuNPs can directly or indirectly detect various specific targets based on different sensing principles, including hydrogen bonds, aptamer-target binding, nucleic acid hybridization, antigen-antibody binding, enzyme inhibition, raman signature, as well as enzyme-mimicking activity [26]. Application of AuNPs in the optical aptasensors for detection of different targets have been discussed in several reviews [5,27,28]. However, the mentioned reviews have discussed about different sensing mechanisms of AuNPs-based biosensors, in this review, we have focused specifically on the latest advancements related to the enzyme-mimicking activity of AuNPs in order to develop design of optical aptasensors and draw attention to the significance of Au nanozymes in the sensing of various analytes.

2. Biosensors and aptasensors

Biosensors are analytical devices that consist of two major parts, a biological detection agent (like antibodies, enzymes, nucleic acids, cell-receptors, microorganism etc.) with another part designed for physical signal transduction component (like optical, and electrochemical) in order to present measurable signals and quantitative analytical information [29]. Recognition elements should demonstrate features, including high sensitivity and selectivity, strong performance and versatility for different targets. Antibodies, enzymes and aptamers are commonly used as recognition elements in the design of biosensors [30]. Aptasensors (aptamer-based biosensors) are biosensors that use single-stranded DNA (ssDNA) or RNA aptamers as their bio-recognition element. Aptamers are short single-stranded (RNA or DNA) oligonucleotides or peptide molecules which are selected by an *in vitro* evolution procedure named SELEX (Systematic Evolution of Ligands by EXponential enrichment) from libraries of random nucleic acid sequences. They exhibit specific binding ability to their targets by folding into three-dimensional structures (such as loop, hairpin and G quadruplex) and also, possess the ability to bind to a large number of target molecules, including proteins, nucleic acids, small molecules, cells and organisms [31–34]. Because of their versatile target binding abilities, aptamers are widely applied in bioanalysis. Compared with receptor-based methods, such as antibodies, aptamers have unique attributes for bioanalytical applications including low-cost and high stability. Moreover, their modification and labeling are easier and they do not have batch to batch variation [35]. All together these features have made aptamers an alternative for antibodies. Various methods have been applied for converting aptamer-target binding into a detectable signal, including electrochemical, optical and electrochemiluminescence methods. Among these different methods, optical analysis has been widely used owing to its high sensitivity and specificity, low-cost, quick response and detection, and simple operation. Optical aptasensors are classified based on different detection methods, including fluorescence, chemiluminescence (CL), colorimetric, SPR and surface-enhanced Raman scattering (SERS) [36–38]. Colorimetric methods using AuNPs are well-known and have been considered for changing signals into visual colors, which can be detected by the naked eye or portable spectrometer [39]. Fluorescence-based biosensors have also been applied for analytical applications because of their sensitivity and efficiency [40]. Fluorescent aptasensors are divided into two groups, labeled and label-free analysis. High performance is one of the main advantages of the labeled aptasensors. However, the main problem with the labeled aptasensors is their high-cost. In the labeled group, different kinds of the fluorophore and/or chromophore are used to make fluorescence property [41]. Chemiluminescence assay is another optical method that is widely used for the design of aptasensors for detection of numerous analytes. This is due to its simplicity, sensitivity and low-cost. Other than fluorescence and chemiluminescence, there are other optical assays that are not as popular as the above-mentioned, for example localized surface plasmon resonance (LSPR) and SERS. One of the most important factors that can increase the sensitivity of biosensors is the use of nanomaterials. Among them, AuNPs are extensively regarded as ideal materials for development of optical sensors [42]. AuNPs can simply be conjugated to different ligands (such as aptamer and antibodies) due to their large specific surface area. Recently, AuNPs were disclosed to be fruitful as artificial enzymes that have inherent peroxidase-like activity and can catalyze different substances to generate a colored product.

3. Optical aptasensors based on Au nanozymes

In the current research, nanoparticles with enzyme-like catalytic activity, called nanozymes have attracted significant attention. The catalytic property of AuNPs as an artificial enzyme has been used in the design of aptasensors to surpass the disadvantages of biological and conventional artificial enzymes. These drawbacks include low catalytic capability in harsh environmental conditions, low operational stability and the high cost for production, separation, and purification [43,44]. In colorimetric aptasensors that are relying on the catalytic activity of AuNPs, chromogenic substrates such as 4-nitrophenol (4-NP), 3,3′,5,5′-tetramethylbenzidine (TMB), 2,2′-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid) diammonium salt (ABTS) and ortho-phenylenediamine (OPD) are mainly used to make colorimetric output indicators.

3.1. 4-Nitrophenol (4-NP)-based (4-NP) aptasensors

The reduction of 4-NP to 4-aminophenol (4-AP) in the existence of too much $NaBH_4$ has often been applied to observe the catalytic activity of AuNPs [45]. AuNPs can catalyze the reduction of 4-NP as a colorimetric probe (yellow color) to 4-aminophenol as a colorless substance. In this regard, several studies exploited 4-NP in design of their colorimetric aptasensors. In one study, Chen et al. designed an ultrasensitive colorimetric Au-based aptasensor for the determination of thrombin in human serum using aptamer-modified AuNPs (Fig. 1). AuNPs nanozymes that had the ability to mimic nitroreductase activity excellently, could catalyze the substrate 4-NP to produce 4-AP in the presence of $NaBH_4$. Upon the addition of thrombin as target, aptamer-thrombin complex was formed and partially covered the surfaces of AuNPs. Therefore, 4-NP had less connection to the AuNPs surface, leading to

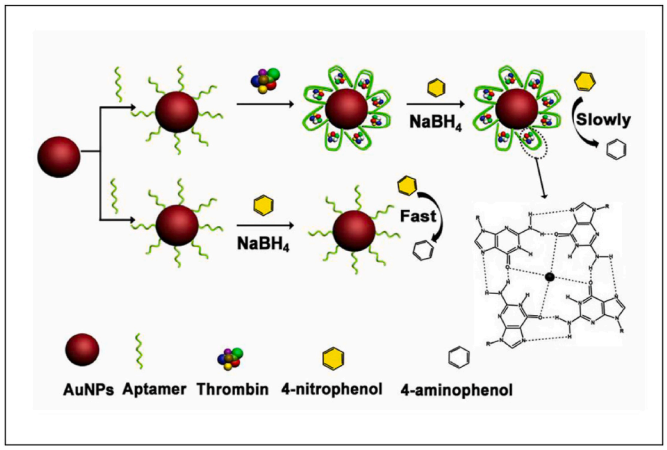

Fig. 1. Schematic representation of the developed aptasensor for detection of thrombin using aptamer-modified AuNPs. Reprinted with permission from Ref. [46].

enhancement of the reduction time of 4-NP to 4-AP caused by the catalytic capability of AuNPs and the color remained yellow. On the contrary, in the lacking of thrombin, 4-NP was easily absorbed on the surface of AuNPs and decreased the reaction time of 4-NP to 4-AP conversion. This resulted in the color change of the samples to colorless. The method was simple and cost-effective with a limit of detection (LOD) of 0.1 nM and linear range from 0.1 to 15 nM [46].

Even though AuNPs has found growing applications in diagnostic methods, their use is still restricted in medicine because of their limited stability in biological environment. Another challenge is the formation of protein corona, when all the surface of AuNPs gets occupied with complex macro biomolecules such as proteins during first few minutes exposing to biological fluids. It is known that protein corona formation alters the original physicochemical properties of the AuNPs and enhances their hydrodynamic diameter. So, it can change the quality of aptasensor. Different approaches for decrease of protein corona formation on the surface of AuNPs includes coating the surface of AuNPs with polyethylene glycol (PEG) chains (PEGylation), Zwitterions, serum dilution and remove of proteins from sample by acetonitrile [47–49]. In our lab, we introduced a colorimetric approach for the detection of fluoroquinolones (ciprofloxacin) according to the intrinsic peroxidase-like activity of AuNPs and the flower-shaped structure of modified AuNPs (Fig. 2). In this work, AuNPs were modified with complementary strands of aptamer (CS1 and CS2) and aptamer was added finally to form a flower shape on the surface of AuNPs. In the absence of ciprofloxacin, the flower-shaped structure of AuNPs is intact and prevents the exposure of 4-NP to AuNPs. As a result, there is no color change and the samples remain yellow. Addition of target, induces binding of aptamer to ciprofloxacin and aptamer leaves the CSs and surface of AuNPs. Therefore, the flower-shaped structure of the modified AuNPs is disassembled and 4-NP owns more connection to the exposed surface of AuNPs, leading to the reduction of 4-NP to 4-AP by the surface of AuNPs. Thus, the color of the sample changes from yellow to colorless which can be observed by the naked eye. This aptasensor showed high selectivity for ciprofloxacin with LOD of 1.3, 2.6 and 3.2 nM in spiked water, serum and milk samples, respectively [50].

In another study, our group fabricated a colorimetric method for the sensitive determination of zearalenone (ZEN) using exonuclease III (Exo III), a non-target-induced aptamer walker and the peroxidase-mimicking activity of AuNPs. Interestingly, without the presence of target, the aptamer can walk on the surface of AuNPs, binds with the CS, and as a result create a dsDNA structure. Upon the addition of Exo III, the complementary strands are digested and the intact aptamer remains on the AuNPs surface. So, 4-NP can simply arrive to the surface of AuNPs and be reduced to 4-AP, leading to the change of color of sample from yellow to colorless. In the presence of the target, the aptamer can bind with ZEN and form a ZEN-aptamer complex, resulting in its separation from its CS. Therefore, there is no dsDNA structure on the surface of AuNPs, and Exo III cannot digest aptamer and CS as ssDNAs. In this situation, 4-NP has less access to the surface of AuNPs owing to the steric impediment from the Aptamer/ZEN complex and intact CS, and the color of 4-NP remaines yellow. The LOD was 10 ng/L. A low concentration of the target requirement and signal enhancement were several of the properties of the fabricated method [51]. Recently, our team developed a new and simple colorimetric aptasensor based on the peroxidase-mimicking performance of AuNPs for the detection of cocaine (Fig. 3). In this study, upon the addition of cocaine, a triple-fragment aptamer (TFA, including; FA1, FA2, FA3) was formed and masked the surface of AuNPs. As a result, the catalytic ability of the surface of AuNPs was prevented. So, yellow 4-NP had less access to the surface of AuNPs, leading to no color change. In the absence of cocaine, AuNPs were modified only with FA1. Thus, the catalytic activity of the surface increased, and 4-NP could easily reach the surface and become colorless (4-AP). This sensing strategy could detect cocaine in a short period (35 min) in spiked human serum with a LOD of 440 pM [52].

In another research, a sensitive colorimetric aptasensor was presented for the determination of potassium (I) in tap water based on the catalytic abilities of AuNPs. In this study, without the presence of target, 4-NP had free access to the AuNPs. This resulted in the increased catalytic activity of the AuNPs surface and as a result 4-NP became colorless. However, in the presence of potassium (I), the aptamer folded into a four-stranded tetraplex structure (G-quartet) with K^+, which prevented the exposure of 4-NP to the catalytic AuNPs surface. Hence, the reduction time of 4-NP by $NaBH_4$ increased. The linear range was 0.1 nM–10 μM with a LOD of about 0.06 nM [53].

3.2. 3,3′,5,5′-tetramethylbenzidine (TMB)-based aptasensors

Nanomaterials such as AuNPs can catalyze the oxidation of the colorless TMB in the existence of H_2O_2 to make a blue product that is utilized as a colorimetric output signal. TMB is positively charged and has two amine groups. This results in its strong affinity toward the negatively charged AuNPs. Several colorimetric aptasensors based on the oxidization of TMB have been reported. In one study by Hu et al. a simple colorimetric aptasensor was proposed for the determination of ricin based on the increased oxidase-like catalytic activity of AuNPs. In the absence of ricin, the aptamer was absorbed onto the AuNPs surface and increased the negative charges of the AuNPs surface, resulting in

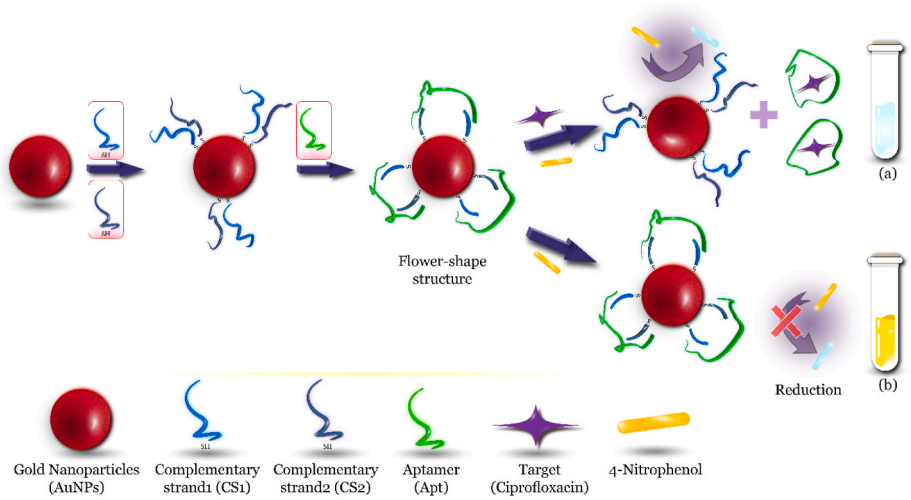

Fig. 2. Schematic representation of the colorimetric aptasensor for determination of ciprofloxacin based on the intrinsic peroxidase-like activity of AuNPs and the flower-shaped structure of modified AuNPs. Adapted with permission from Ref. [50].

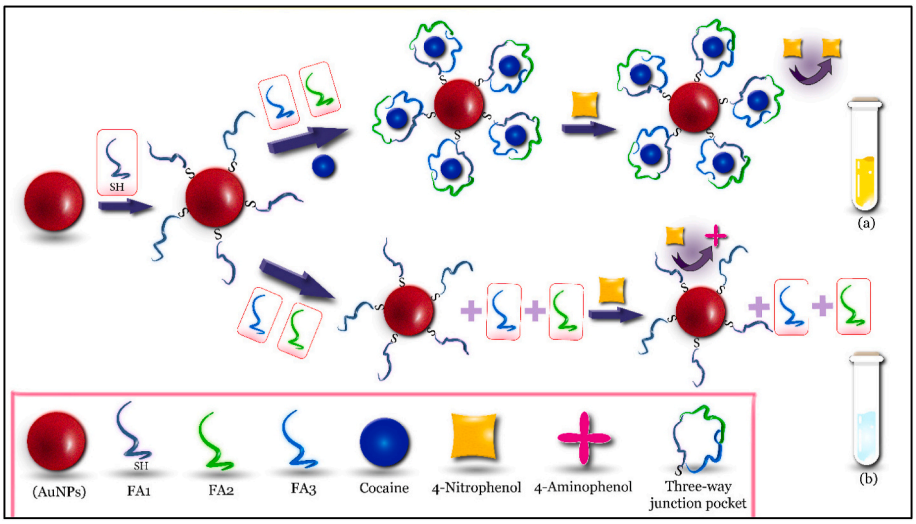

Fig. 3. Illustrations of the sensing mechanism of cocaine using triple-fragment aptamer and peroxidase-mimicking performance of AuNPs. Adapted with permission from Ref. [52].

greater binding affinity with TMB (positive charge). Consequently, the peroxidase-like activity of AuNPs was significantly enhanced. Thus, TMB as a substrate, was oxidized in the presence of H_2O_2, resulting in a color change of the solution. On the contrary, when the target was added, the aptamer could bind with ricin to form an aptamer-ricin complex which was released from the surface of AuNPs. Consequently, the negative charge of the surface became less, indicating a drop in the catalytic activity of AuNPs. The color change from dark-blue to light-blue was observed which was measured by a UV–vis spectrometer or by using the naked eye. The designed colorimetric approach has a linear range from 0.05 nM to 10 nM with a LOD of 0.05 nM [54].

In another study, Hu et al. developed a AuNPs-based aptasensor for the detection of abrin in raw milk with a similar strategy. In the presence of target, the catalytic activity of AuNPs led to a switch in the color of the sample from dark-blue to light-blue. The developed method was sensitive, simple and inexpensive with a LOD of 0.05 nM [55].

A new, rapid and ultrasensitive colorimetric assay based on aptamer and peroxidase-like activity of AuNPs was reported for the determination of murine norovirus (MNV). Upon addition of norovirus, the aptamer was attached to its target and left the surface of the AuNPs, leading to an increase in the catalytic ability of AuNPs. The sample color changed to blue through the oxidation of a colorless peroxidase substrate (TMB). Whereas without norovirus in the sample, but in the presence of other contaminants (*Staphylococcus aureus, Escherichia coli)*, the aptamers had no affinity to bind with other non-specific targets and thus, remained on the AuNPs surface. So, no catalytic signal enhancement of AuNPs was exhibited. TMB was not oxidized and the solution showed no-color. The detection limit was just 20 viruses per assay equivalent to 200 viruses/mL. This aptasensor simply can detect norovirus with high sensitivity and reproducibility, without need of sophisticated infrastructure [56].

Yan et al. employed a colorimetric biosensor for the determination of sulfadimethoxine, applying a DNA aptamer and AuNPs. In the lack of target, the aptamer remains on the AuNPs surface and restrains the peroxidase-like activity of AuNPs. However, in the presence of sulfadimethoxine, the aptamer has a higher binding constant for the target than AuNPs, so it desorbs from the surface of AuNPs. Thus, the catalytic ability of the AuNPs is reactivated, which can catalyze a colorless TMB to a purplish-blue oxidized product. This process can be monitored by a UV–visible absorptiometry at 650 nm. The proposed aptasensor can detect sulfadimethoxine with a LOD of 10 ng/mL [57]. In another interesting study, a simple and feasible colorimetric aptasensor with high sensitivity was introduced by Chen et al. for detection of K^+. The peroxidase-mimicking of AuNPs and K^+ specific binding aptamer were used in this design. The presence of aptamers on the surface of AuNPs significantly improved the peroxidase-like activity of AuNPs because of the increased affinity between the aptamer-modified AuNPs and TMB., In the presence of the target, aptamer interacted with K^+ and formed a G-quadruplex structure, indicating less peroxidase-like activity of AuNPs and so, observation of light-blue color in the sample. In the absence of K^+, aptamer remained on the surface of AuNPs. Thus, TMB was more oxidized in the presence of H_2O_2 by the AuNPs and the solution showed a dark-blue color. The color change of solution is related to the amount of target which can be measured by a UV–vis spectrometer or even by the naked eye. Concentrations as low as 0.06 nM K^+ could be measured by this aptasensor [58]. Similarly, Zhang and Li. prepared horseradish peroxidase (HRP) functionalized AuNPs for the colorimetric detection of thrombin in spiked human serum at very low concentrations. Without the presence of target, the 15-mer thrombin binding aptamer (TBA) enhanced the catalytic activity of AuNPs to oxidase TMB and yield a dark-blue color. Upon addition of thrombin, TBA was detached from the surface of AuNPs, which led to a decrease in the peroxidase-like activity of AuNPs and so, a reduction of the oxidation of TMB in the presence of H_2O_2. The data proved that the aptasensor has high selectivity and sensitivity for the detection of thrombin with the LOD of 0.02 pM and a linear range from 0.1 pM to 0.1 μM [59].

A new colorimetric method based on peroxidase-like nanozyme activity of AuNPs was reported for the highly sensitive recognition of a pesticide (acetamiprid) (Fig. 4). When the target is absent from the system, the acetamiprid-specific S-18 aptamer completely masks the AuNPs surface, resulting in the inhibition of the intrinsic catalytic

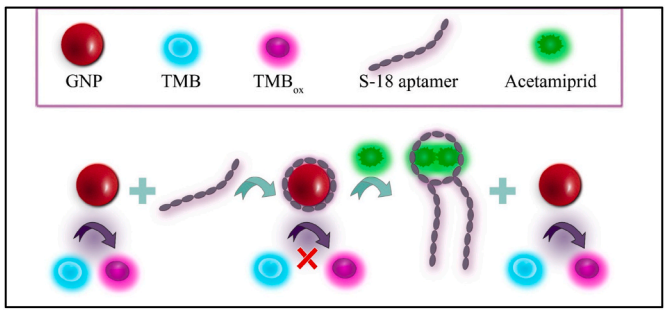

Fig. 4. Illustrations of the sensing mechanism of acetamiprid (Adapted with permission from Ref. [60].

activity of AuNPs. Thus, the reaction product remains colorless. In the presence of acetamiprid, upon conjugation of target and S-18 aptamer, the color of the solution changes to purplish-blue. This color change is due to the catalytic activity of AuNPs which enables the oxidation of TMB in the presence of H_2O_2. This proposed sensor exhibited high selectivity and sensitivity with a detection limit of 0.1 ppm and a dynamic linear range of 0.1–10 ppm [60].

It has been proved that an additional surface coating or modification of AuNPs nanozymes would affect the enzymatic activities of AuNPs via the alteration of surface charge and the shielding of active sites. Generally, the extra coating would shield and protect the active sites and thus, reduces the catalytic activities of AuNPs [61]. For instance, in a colorimetric aptasensor, cetyltrimethylammonium bromide (CTAB) could effectively decrease the oxidation of TMB in the presence of AuNPs nanozymes and H_2O_2 when the target (Malachite Green) was present in the sample. In this situation, aptamer bound to MG and so, CTAB could aggregate AuNPs and reduce the enzymatic activities of AuNPs [62]. Using this phenomenon, as low as 1.8 nM MG was detected in aquaculture water by the colorimetric method in the concentration range from 10 to 500 nmol L^{-1} [63].

3.3. 2,2-Azino-bis (3-ethylbenzothiazoline-6-sulfonic acid) (ABTS)-based aptasensors

2,2-azino-bis (3-ethylbenzothiazoline-6-sulfonic acid) ABTS has a negative charge and is considered as a common signal reporter with a green oxidation product and a typical absorption peak at 733 nm. This reaction is catalyzed by AuNPs in the presence of hydrogen peroxide [64]. Yang et al. investigated a new, convenient and sensitive colorimetric assay for the determination of acetamiprid in wastewater and tomatoes. It was based on the S-18 (aptamer)-target specific recognition, ABTS as a colorimetric probe and the peroxidase-like catalytic activity of AuNPs. Briefly, upon the addition of acetamiprid to the sample, the aptamer-target complex was formed. Thus, the peroxidase activity of the AuNPs remained intact which enabled AuNPs to oxidize ABTS to yield green products. Conversely, in the lack of the target, the aptamer was adsorbed on the surface of AuNPs and repeled ABTS due to shielding effect of the negatively charged aptamer. Thus, the color of the solution remained red. The prepared aptasensor was able to detect acetamiprid with LOD as low as 1.02 μg L^{-1} and a dynamic range of 10–160 μg L^{-1} [65].

Similar to this strategy, Zhao group introduced a colorimetric aptasensor for the determination of streptomycin (STR) as an antibiotic, using DNA aptamer (STR1-aptamer) and AuNPs. In the absence of the target, AuNPs surface is still masked with the STR1-aptamer, which decreases the catalytic activity of AuNPs. Thus, AuNPs cannot catalyze the oxidation of ABTS by H_2O_2 and no color change is observed in the sensing solution. After the addition of STR, the STR-STR1 complex is formed and leaves the surface of AuNPs, improving the ability of AuNPs to catalyze ABTS in the presence of H_2O_2. This results in the color change of the sample to green, which was measured by the UV/vis spectrophotometer. The main benefit of the proposed aptasensor was the precision and reproducibility with LOD as low as 86 nM [66]. However, the main challenge in aptasensors based on ABTS is the oxidation product of ABTS which is not stable and can rapidly decay to colorless in solution [67].

3.4. Luminol–H_2O_2–based aptasensors

Chemiluminescence (CL)-based methods have been used because of their sensitivity and wide linear quantitative range of detection. These detection methods are based on the achievement of signals from the luminol-H_2O_2 reaction [68]. Shwetha and coworkers, designed a CL-based detection system using AuNPs for the detection of a tumor suppressor protein (p53) (Fig. 5). In the absence of p53, the aptamer still remains on the AuNPs surface and can protect the AuNPs against

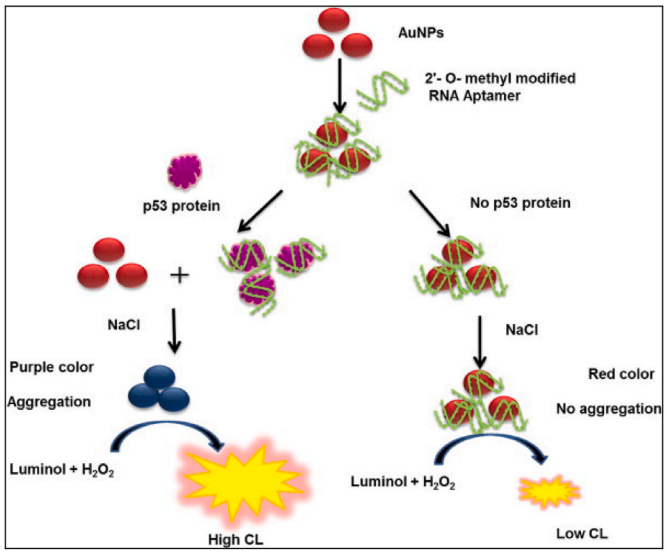

Fig. 5. Scheme of the aptasensor for P53 protein determination using AuNPs and luminescent signal. Reprinted with permission from Ref. [69].

salt-induced aggregation. Therefore, the dispersed AuNPs generate a weak luminescent signal. However, in the presence of p53, the p53 aptamer is converted from a coil structure to a G-quadruplex structure in order to bind to its target. So, the addition of salt leads to aggregation of AuNPs and the color of the solution changes from red to purple (LOD of 0.1 ng/mL for the colorimetric aptasensor) and induced a strong CL signal for the luminol-H_2O_2 system (LOD of 10 pg/mL based on CL detection) upon the reaction of luminol-H_2O_2 with the aggregated AuNPs [69].

In another work, a sensitive and a label-free aptasensor was presented for the recognition of thrombin based on the catalytic activity of AuNPs on the luminol-H_2O_2 CL response. In the absence of the thrombin, ssDNA aptamer could be adsorbed onto the AuNPs surface and prevent the aggregation of AuNPs by salt, leading to the production of a weak CL signal. Upon the addition of thrombin, aptamer was folded into a three-dimensional structure and attached to its target and had no interaction with AuNPs. So, the addition of salt, induced aggregation of AuNPs that increased the catalytic ability of the AuNPs on the luminol-H_2O_2 CL response. These aggregated AuNPs showed a strong CL signal. The obtained LOD was as low as 26 fM [70].

Qi and co-workers introduced a CL method for recognition of bisphenol A (BPA), a ubiquitous pollutant, in dismantling site of electronic waste using positive AuNPs CL system and ssDNA aptamer (Fig. 6). In this method, the detection of BPA by an anti-BPA aptamer in the environmental medium can be achieved through the catalytic ability of cationic AuNPs in the luminol-$AgNO_3$ CL reaction. When BPA is absent from the system, the anti-BPA aptamer as a ssDNA structure can bind on the positive surface of AuNPs, leading to the protection of the

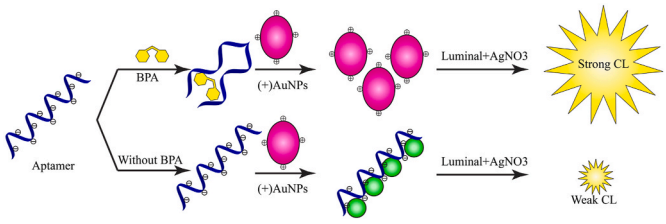

Fig. 6. Schematic illustration of the CL sensing mechanism for detecting BPA using aptamer and AuNPs. Reproduced with modification and permission from Ref. [71].

electropositive surface of AuNPs. Therefore, AuNPs cannot interact with luminol (anions). As a result, the concentration of substances decreases and generates a weak luminescent signal. Upon addition of BPA, the anti-BPA aptamer changes its conformation to bind with the target and cannot easily wrap around the cationic AuNPs that leads to a powerful CL signal in the attendance of luminal and $AgNO_3$. Applying this method, the LOD of 6.2×10^{-2} ng/mL was achieved that was less than the requirements of Chinese Health Standards (0.05 mg/kg) [71].

3.5. Ortho-phenylenediamine (oPD)-based aptasensor

Jeon et al. described a fast and washing free colorimetric method based on AuNPs for detection of soluble interleukin-2 receptor a (sIL-2Ra). sIL-2Ra is a useful biomarker for disease activity in different inflammatory diseases. When the target is absent, the sIL-2Ra aptamer is attached onto the surface of the AuNPs and increases the negative charge of their surface. Then, the AuNPs catalyze the oxidation of oPD as a positively charged peroxidase substrate in the attendance of H_2O_2 and make deeper brown color. Even so, in the existence of sIL-2Ra, the aptamers is desorbed from AuNPs surface and reduce the catalytic activity of AuNPs, resulting in less color development. The LOD of this aptasensor was 1 nM in both buffer and spiked human serum with a linear range of 1–100 nM [72]. All the mentioned methods were summarized in Table 1.

4. Conclusion and future perspectives

The enzyme mimetic property of AuNPs whit various surface modifications have facilitated their usage in biological applications. They possess some advantages, including low-price, high stability against denaturing, easy preparation and synthesis. In this way, optical aptasensors especially colorimetric aptasensors based on the outstanding catalytic activity of AuNPs have gained a great deal of attention [42,73]. AuNPs-based aptasensors with the characteristics of flexibility, high sensitivity and selectivity, simple operation, inexpensive, reproducible and fast response can be applied broadly for multipurpose settings. Reproducibility of this type of aptasensor enabling more simply fabrication processes by different researchers [74]. Moreover, the optic results can be visualized or quantified by monitoring color alteration simply using naked eye or spectrophotometer. From the sensing view, the intrinsic properties of AuNPs, including the combination of colloidal stability, low toxicity, compatibility to aqueous medium, high surface area, easy surface functionalization along with miniaturization, allow them ideal candidates to be used in the design of smart sensors for detection of target samples [75]. In addition, the presence of AuNPs in the construction of aptasensors augments the signal of many transducers. Even though notable advances haven been attained in AuNPs-based aptasensors, some drawbacks and problems exist that should be further addressed. Firstly, the color change from red to blue of AuNPs is hard to observe by naked eye in some cases particularly at a low concentration of analyte. So, Signal amplification approaches should be consider to improve the detection sensitivity for naked eye readout. Another important issue is the stability of the AuNPs-based sensor which should be improved for quantitative detection. Thirdly, only limited aptamers against specific analytes have been investigated and selected, thus additional more aptamers for designing various aptasensors are needed to identify [67,76]. In this review, we have focused on the recent achievements of optical aptasensors specifically colorimetric sensing methods combined with the visual signals induced by the catalytic activity of AuNPs. Aptasensors using catalytic activity of AuNPs are generally promising methods for on-site and real-time detection of different targets without the aid of any advanced instrument. We believe that in the near future, there will be a tremendous growth in the development of AuNPs-based aptasensors for diagnostic applications providing that the drawbacks of AuNPs like corona shield effect are addressed well.

Table 1
Optical aptasensors based on the catalytic activity of AuNPs.

Analytical technique	Strategy	Target	Sample	LOD	Linear range	Ref
Colorimetry	4-nitrophenol	Thrombin	Human serum	0.1 nM	0.1–15 nM	[46]
Colorimetry	4-nitrophenol	Ciprofloxacin	Spiked water, serum and milk	1.3 (spiked water), 2.6 (serum)and 3.2 (milk) nM	4 nM–500 nM	[50]
Colorimetry	4-nitrophenol	Zearalenone	Human serum	10 ng/L	20–80000 ng/L	[51]
Colorimetry	4-nitrophenol	Cocaine	Spiked human serum	440 pM	Over 2–100 nM.	[52]
Colorimetry	4-nitrophenol	K(I)	Urine	0.06 nM	0.1 nM to1 μM	[53]
Colorimetry	3,3,5,5-tetramethylbenzidine (TMB)	Ricin	Raw milk and Pepsi Cola	0.05 nM	0.05 nM–10 nM	[54]
Colorimetry	3,3,5,5-tetramethylbenzidine (TMB)	Abrin	Raw milk	0.05 nM	0.2 nM–17.5 nM	[55]
Colorimetry	3,3,5,5-tetramethylbenzidine (TMB)	Murine norovirus (MNV)	Viral sample	200 viruses/mL		[56]
Colorimetry	3,3,5,5-tetramethylbenzidine (TMB)	Sulfadimethoxine	Milk	10 ng·mL−1	(0.01–1000 μg·mL−1)	[57]
Colorimetry	3,3,5,5-tetramethylbenzidine (TMB)	K	Tap water	0.06 nM	0.1 nM to 10 μM	[58]
Colorimetry	3,3,5,5-tetramethylbenzidine (TMB)	Thrombin	Human serum	0.02 pM	0.1 pM-0.1 μM	[59]
Colorimetry	2,2-azino-bis (3-ethylbenzothiazoline-6-sulfonic acid) ABTS	Acetamiprid	Environmental samples	0.1 ppm	0.1–10 ppm	[60]
Colorimetry	3,3,5,5-tetramethylbenzidine (TMB)	Malachite Green (MG)	Aquaculture water	1.8 nmol L^{-1}	10–500 nmol L^{-1}	[63]
Colorimetry	3,3,5,5-tetramethylbenzidine (TMB)	Acetamiprid	Waste water and tomatoes	1.02 μg L^{-1}	10–160 μg L^{-1}	[65]
Colorimetry	2,2-azino-bis (3-ethylbenzothiazoline-6-sulfonic acid) ABTS	Streptomycin	Milk	86 nM	0.1 μM–0.5 μM	[66]
Colorimetry/ Chemiluminescence	Aggregation of AuNPs by salt and catalytic activity of gold-nanoparticle	P53	Serum	0.1 ng/mL for colorimetric assay and 10 pg/mL for CL assay	10 pg/mL to 1 ng/mL	[69]
Chemiluminescence	Aggregation of AuNPs by salt and catalytic activity of gold-nanoparticle	Thrombin	Bovine serum albomin	26 fM	From 6.3 to 10^{-14} to 6.3-10^{-11}M	[70]
Chemiluminescence	Catalytic ability of cationic AuNPs in luminol-AgNO3 CL reaction	BPA	Dismantling site of E-waste	6.2×10^{-2} ng/mL		[71]
Colorimetry	oxidation of ortho-phenylenediamine (oPD)	Interleukin-2 receptor a (sIL-2Ra)	spiked human serum	1 nm	1–100 nm	[72]

Conflict of interest

There is no conflict of interest in this article.

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