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RESEARCH ARTICLE

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Chemiluminescence assay for kanamycin based on target recycling strategy

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Abstract

A chemiluminescence (CL) sensing strategy for kanamycin residue detection in fish samples was established based on luminol-functionalized gold nanoparticles as CL nanoprobe materials combined with DNA hairpin structure and carboxyl-modified magnetic beads. Relying on nucleic acid amplification technology, the system can successfully realize the recycling of kanamycin, so that the biosensor can release a large number of luminol-functionalized gold nanoparticles with excellent CL performance even at a low residual levels of kanamycin. The biosensor strategy showed a good linear relationship with kanamycin in the range 0.09–130 nM, the detection limit was as low as 0.04 nM. This method proves the excellent performance of the sensing strategy and provides a low-cost and high-sensitivity CL analysis strategy for the detection of kanamycin and even other antibiotics.

KEYWORDS
antibiotics, aptamer, AuNPs, chemiluminescence, recycling

1 | INTRODUCTION

Kanamycin is a type of aminoglycoside antibiotic that has been widely used in animal husbandry and the food industry because of its excellent efficacy. The correct use of kanamycin can effectively prevent infection problems in animal husbandry, fishery, etc.[1] In contrast, excessive use of kanamycin can lead to excessive concentrations of kanamycin in daily food such as fish and milk, which poses a threat to human health and the public environment.[2] For this reason, the European Union, China and other regions have issued strict standards to stipulate the maximum allowable residue limit (MRL) of kanamycin in food. For China, the MRL of kanamycin in milk is 200 μg kg^{-1}, while in Europe the limit is 150 μg kg^{-1}.[3] The current detection methods such as gas chromatography, high performance liquid chromatography (HPLC),[4–6] which have been listed as standard methods in most countries and regions, are limited by some shortcomings, such as the inability to quickly test in the field, complicated operation, and long test cycle, etc. Although new methods such as colorimetry[7] and electrochemistry[8] can achieve high-sensitivity detection of low-level kanamycin, they are susceptible to external interference and the electrode preparation is relatively complicated. There is an urgent need for a new strategy to quickly and easily detect the level of kanamycin residues in samples.

Aptamers are single-stranded DNA or RNA oligonucleotides with high specificity and affinity for target substances screened from random oligonucleotide libraries based on Systematic Evolution of Ligands by EXponential enrichment (SELEX).[9–11] As an excellent tool of biological science, the aptamer has been widely used in the detection of cells, viruses, proteins, toxins, allergens, and other biological components because of its high affinity, strong specificity, large surface area, a large number of receptor binding sites, and a wide range of targets.[12–14] Catalytic hairpin assemblies (CHA), as a

mature enzyme-free DNA amplification technique with high efficiency and target cycle, have been applied in the field of signal transduction and amplification related to nucleic acid analysis and biosensor.[15,16]

Chemiluminescence (CL) is a phenomenon that produces light radiation through chemical reactions.[17,18] It has the strengths of high sensitivity, wide linear range, low detection limit, and no need for an excitation light source. As an efficient and simple analytical method, it has had brilliant performance in the fields of biosensor, clinical medicine, and chemical detection.[19–21] Luminol and its derivatives, as one of the classical luminescent reagents used in CL analysis, are widely used in CL analysis and detection because of relatively good stability, relatively high luminescence quantum yield, simple synthesis method, and other characteristics.[22] Cui's group[23] successfully achieved direct reduction of $HAuCl_4$ with luminol and obtained gold nanoparticles coated with multiple luminol molecules and their oxidation products, which not only had outstanding CL properties, but also showed satisfactory catalytic performance and biocompatibility. The ability to bind sulfhydryl-functionalized stem–loop DNA molecules makes luminol-functionalized gold nanoparticles (LumAuNPs) one of the choices for CL probe materials.

In this work, the CL method was used to detect the kanamycin residual level. By combining aptamer and CHA technology, two different hairpins were used to achieve target circulation and signal amplification. LumAuNPs with excellent CL performance and catalytic performance were used as probes to reflect the kanamycin content in the sample. Based on target circulation and CHA technology, a simple, fast, low-cost, and high-sensitivity CL analysis strategy for kanamycin detection was satisfactorily realized.

2 | EXPERIMENTAL

2.1 | Regent and materials

Kanamycin, tris(2-carboxyethyl)phosphine hydrochloride (TCEP), luminol, and tris-(hydroxymethyl)aminomethane (Tris) were obtained from Aladdin (Shanghai, China). Chloroauric acid ($HAuCl_4$) was purchased from Sigma-Aldrich (MO, USA). Hydrogen peroxide (H_2O_2) was obtained from Sinopharm Chemical Reagent Co., Ltd (Shanghai, China). 3-Ethylcarbodiimide hydrochloride (EDC) and *N*-hydroxysuccinimide (NHS) were both obtained from J&K Chemical Co., Ltd (Beijing, China). Carboxyl-modified magnetic beads (MBs) was purchased from BaseLine Chromtech Research Center (Tianjin, China). All other reagents were of analytical grade and could be used without further purification. Tertiary distilled water was used throughout the research process.

Phosphate-buffered saline (PBS) (pH = 7.4) was prepared by dissolving 5.93 g NaH_2PO_4, 58 g Na_2HPO_4, and 9 g NaCl in 1000 ml ultrapure water.

All DNA oligonucleotides sequences were synthesized using Shanghai Sangon Biological Engineering Technology Co. Ltd (Shanghai, China) and the sequences are shown in Table S1.

2.2 | Instruments

Transmission electron microscope (TEM) (JEM-2100, JEOL, Tokyo, Japan). The static injection method was used for CL detection, and the CL intensity was measured and recorded using an FI-CL instrument (IFFM-E, Remex Analytical Instrument Co. Ltd, Xian, China). Tertiary distilled water was purified using a purification system (Shanghai Yarong Biochemistry Instrument Factory Co., Ltd, China). Ultraviolet (UV)–visible spectra were acquired with a UV-3600 spectrophotometer (Shimadzu, Tokyo, Japan). All fluorescence data were obtained from an FL-2700 fluorescence spectrophotometer (Shimadzu, Tokyo, Japan)

2.3 | Synthesis of LumAuNPs

LumAuNPs were synthesized by referring to the method of Cui *et al.*[23] All glassware used in the synthesis were cleaned using immersion in aqua regia, then washed thoroughly with ultrapure water, and dried thoroughly before the reaction. LumAuNPs were obtained by direct reduction of $HAuCl_4$ solution with luminol. The specific operation was to put 50 ml of 0.02% (w/w) $HAuCl_4$ solution into a round-bottomed flask and heat it to boiling. Then 1.55 ml of 1.0×10^{-2} M luminol was quickly added under intense stirring and the reaction continued for 40 min. The reaction solution was observed to change from pale yellow to pale black at first. After the reaction solution finally turned purple or wine red, the heating was stopped, and the solution was continued to be stirred to room temperature. Then it was stored at 4°C for later use.

2.4 | Preparation of LumAuNPs conjugated sDNA (LumAuNPs–sDNA)

LumAuNPs–sDNA were made specifically by first adding 5 μl of 10 mM TCEP to 100 μl of 1.0×10^{-6} M sDNA, and shaking for 1 h at 37°C. Then 600 μl of LumAuNPs synthesized before were added to the activated solution, and shaken overnight in a shaker at 37°C. After this process, the LumAuNPs–sDNA were aged for 48 h with 10 mM Tris−HCl (pH = 8.2) containing 0.3 M NaCl. The red precipitate appeared after centrifugation at 12,000 rpm for 10 min. After the supernatant was removed, the red precipitate was washed with 0.1 M PBS and centrifuged in the same operation. After three times, the red precipitate was finally dispersed into 1000 μl of PBS and refrigerated for further use.

2.5 | Preparation of MB conjugated HP1 (MB-HP1)

First, 200 μl of 5.0×10^{-8} M HP1 was heated in a 95°C water bath for 5 min, and then cooled to room temperature naturally to form a stable hairpin structure. Next, 50 μl carboxyl-modified MBs were

purified by 0.1 M PBS (pH = 7.0) for three times in a centrifuge tube. The washed MBs were dispersed by 1000 μl 0.1 M PBS containing 0.1 M EDC and 0.05 M NHS. The centrifuge tube was put in a shaker at 37°C for 30 min. After the reaction, annealed HP1 was added to the centrifugation tube and shaken overnight at 37°C to obtain MB-HP1. The obtained MB-HP1 was magnetically separated, washed three times with 200 μl 0.1 M PBS (pH = 7.4), then dispersed into 200 μl 0.1 M PBS (pH = 7.4) and stored at 4°C.

2.6 | Preparation of MB-HP1 conjugated LumAuNPs–sDNA

Before magnetic separation, 100 μl MB-HP1 and 100 μl LumAuNPs–sDNA were incubated at 37°C for 2 h to form MB-HP1-sDNA-LumAuNPs. The formed MB-HP1-sDNA-LumAuNPs was rinsed three times with 0.1 M PBS (pH = 7.4), then dispersed in 200 μl 0.1 M PBS (pH = 7.4) and stored at 4°C.

2.7 | CL detection of kanamycin

To determine kanamycin, 50 μl MB-HP1-sDNA-LumAuNPs were mixed with different concentrations of kanamycin and incubated for 30 min. The mixed solution in the centrifuge tube was added to 10 μl 10 μM HP2 and incubated at 37°C for 30 min. After magnetic separation, the supernatant was injected into H_2O_2 and the CL signal was recorded.

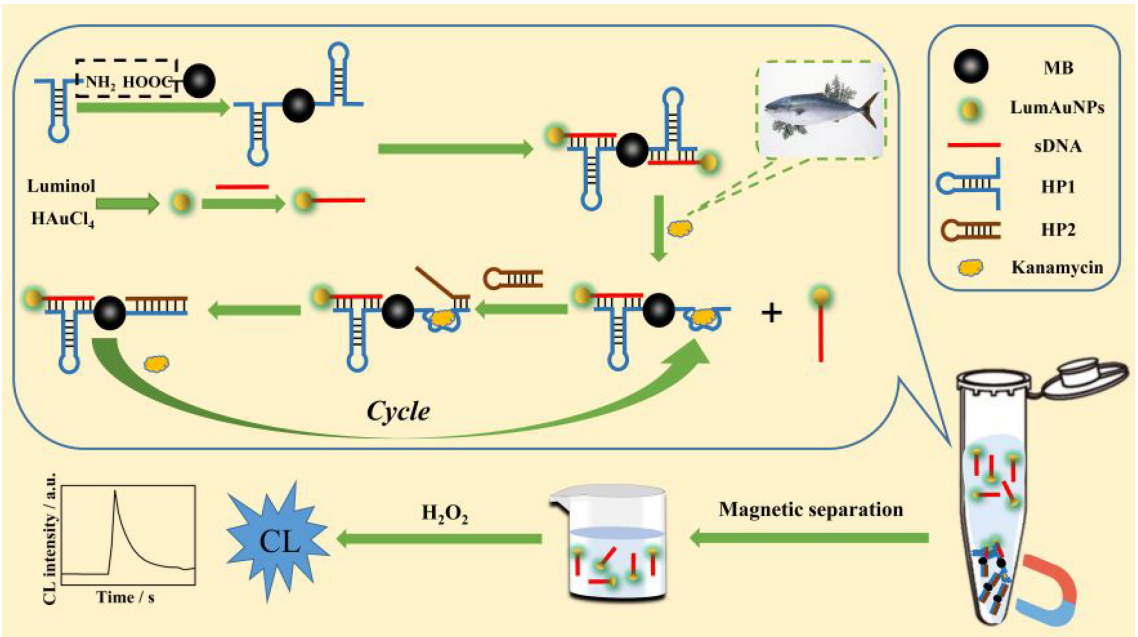
2.8 | Preparation of real samples

The live fish samples were purchased from the local seafood market in Qingdao, and kanamycin was not used in the breeding place of the purchased fish during the breeding process. Then the fish was processed. We weighed ~3.0 g of fresh fish sample, placed it in a centrifuge tube and ground it, added 6.0 ml of 3% (W/V) trichloroacetic acid aqueous solution, then added 3.0 ml of methanol to shake well, and then the centrifuge tube was centrifuged for 10 min, the supernatant was taken and dried under a nitrogen atmosphere, the obtained solid was dissolved in PBS (pH = 7.4), and stored under refrigeration.

3 | RESULTS AND DISCUSSION

3.1 | Principles of kanamycin detection

LumAuNPs combined with sDNA were oxidized using H_2O_2 to generate a CL signal to trigger the kanamycin detection system as shown in Scheme 1. The hairpin structure HP1 containing the kanamycin aptamer was combined with carboxyl-modified MBs through amidation reaction between carboxyl group of MBs and amino of HP1.[24] Relying on base complementation, the sDNA labelled with LumAuNPs was linked with MB-HP1 to form MB-HP1-sDNA-LumAuNPs that could specifically recognize kanamycin. When kanamycin was present in the system, it was accurately recognized by MB-HP1-sDNA-LumAuNPs, the hairpin structure HP1 containing the kanamycin aptamer sequence bound to kanamycin



SCHEME 1 Illustration of CL biosensor strategy for kanamycin detection

and LumAuNPs–sDNA which could generate CL signals were released. After the addition of the hairpin structure HP2, the exposed end of MB-HP1 bound to kanamycin and the exposed end of HP2 began to complement each other until they were completely complementary. Kanamycin, which was previously bound to MB-HP1, was then released into the reaction solution, allowing kanamycin to re-enter the circulation. The kanamycin re-entered into the new cycle was recognized and bound again, so that another LumAuNPs–sDNA was released. The repeated release of LumAuNPs–sDNA increased the amount of luminescent substances in the solution, which resulted in the acquisition of a strong CL signal. This process allowed kanamycin to be recycled, and more CL probe LumAuNPs–sDNA were released, therefore realizing the result that kanamycin at low concentration levels can lead to the release of a large amount of LumAuNPs–sDNA, After magnetic separation, the supernatant in the centrifuge tube was reacted with H_2O_2, and finally an amplified CL signal was generated. When HP2 was not added to the system, kanamycin could not be recycled, and the CL intensity that the system could generate was limited. Compared with the CL intensity that the system could produce after adding HP2, the sensitivity without adding HP2 was relatively poor.

3.2 | Characterization of the materials used

The morphologies of LumAuNPs and MB-HP1-sDNA-LumAuNPs were characterized at different magnifications using TEM, UV–visible spectra, and fluorescence spectra were used to prove the successful synthesis of the materials used.

When the reaction solution turned wine red or purple, LumAuNPs were successfully synthesized. As clearly shown in the TEM image (Figure 1a), it can be seen that the synthesized LumAuNPs had an approximate spherical object with an average size of 30 nm. In addition, in the UV–visible absorption spectrum of Figure 1c, it can be seen that the prepared LumAuNPs had a clear UV absorption peak at 520 nm compared with luminol, gold nanoparticles (AuNPs) and LumAuNPs–sDNA. It also had all the characteristic peaks of luminol, indicating that LumAuNPs were successfully synthesized. It can be clearly seen from the Figure 1c that, compared with LumAuNPs, LumAuNPs–sDNA has an absorption peak at 260 nm, which proves that sDNA was successfully bound to LumAuNPs. As can be seen from Figure 1d, the synthesized LumAuNPs had fluorescence properties and the CL peak appeared at ~425 nm, which can prove that the LumAuNPs were well synthesized and successfully linked to the mercapto-modified sDNA through the Au–S bond.

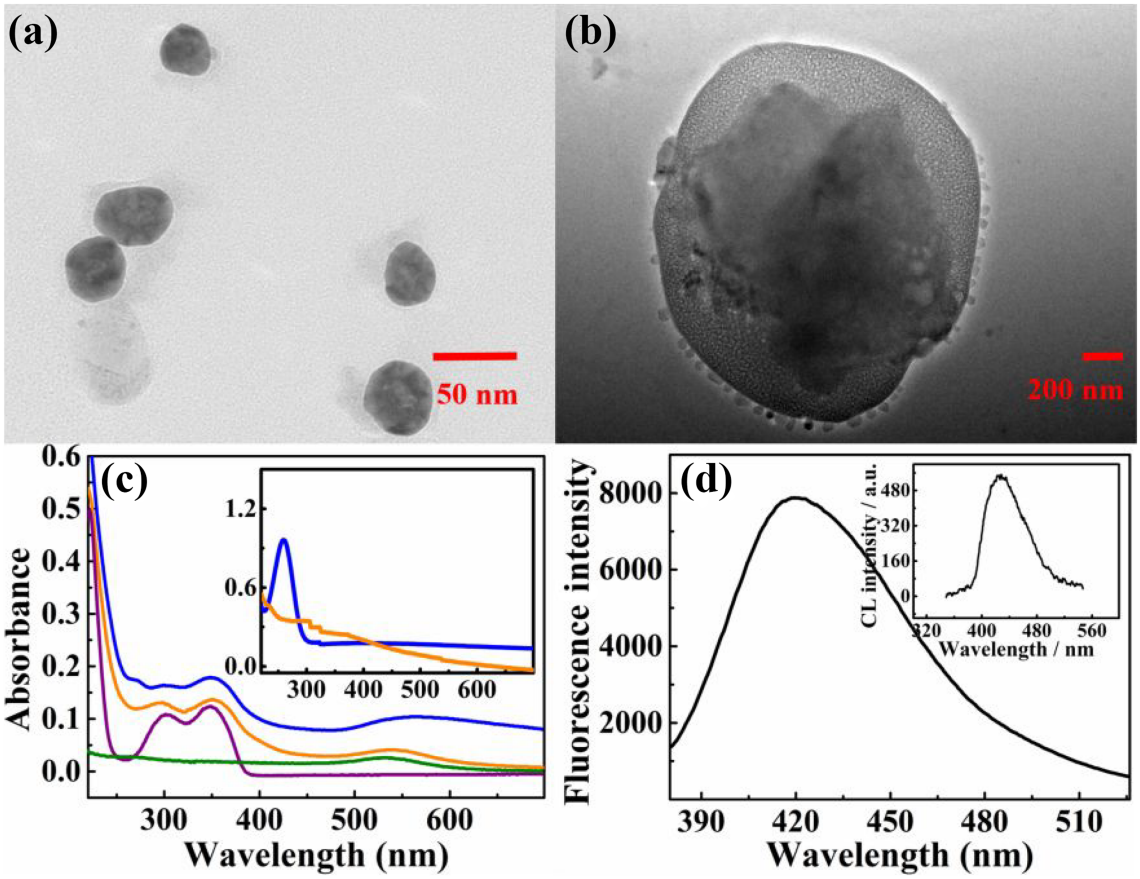


FIGURE 1 Transmission electron micrographs of LumAuNPs (a) and LumAuNPs combined with magnetic beads (MB) (b). (c) UV–visible spectra of luminol (purple), AuNPs (green), LumAuNPs (orange), LumAuNPs–sDNA (blue). Inset: MB (Orange) and MB-HP1 (blue). (d) Fluorescence spectra of LumAuNPs and CL spectra of LumAuNPs-H_2O_2

The UV–visible spectrum of Figure 1c clearly shows that the MBs amidated with HP1 have a strong UV absorption peak at 260 nm, which proves that there is a strong interaction between the amino-containing HP1 and carboxy-MBs, and that HP1 had successfully attached to the MBs. Figure 1b is a TEM image reflecting MB-HP1-sDNA-LumAuNPs. As can be clearly seen from Figure 1b, large numbers of LumAuNPs with an average diameter of 30 nm were assembled on the surface of MBs. After incubation for a period of time, the nanoprobe was successfully combined with the MBs, which proves that the synthesized MB–HP1–sDNA–LumAuNPs were satisfactory. At this time, MB–HP1–sDNA–LumAuNPs had become a biosensor with good specificity and excellent CL performance.

3.3 | Cycle verification of the nucleic acid used

Polyacrylamide gel electrophoresis (PAGE) analysis was used to verify the success of the target cycle used. The six lanes designed (Figure 2) clearly show the success of the kanamycin cycle. First, the first three lanes were separate HP1, sDNA, and HP2. The fourth channel was HP1 and sDNA. It can be seen that the band appearing in the fourth lane was above the first three lanes, which proves that HP1 and sDNA have successfully combined into a whole. Kanamycin was added to the next lane. The addition of kanamycin destroyed the previous HP1-sDNA, and HP1 combined with kanamycin instead. HP2 was added to the last channel, HP2 and HP1 were fully complementary and bound as a whole, shown as the brightest bands above the lane. This result fully demonstrated that the kanamycin cycle was successfully triggered in the presence of HP2 and kanamycin.

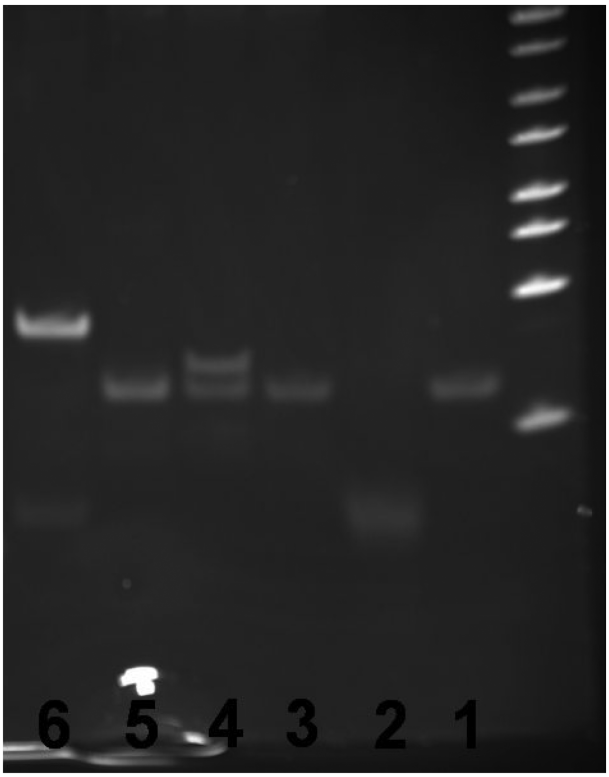


FIGURE 2 Polyacrylamide gel electrophoresis analysis of kanamycin cycling. (1) HP1, (2) sDNA, (3) HP2, (4) HP1 and sDNA, (5) HP1, sDNA and kanamycin, (6) HP1, sDNA, HP2, and kanamycin

3.4 | Optimization of experiment conditions

It is well known that the temperature of incubating DNA has an important effect on the detection performance of the biosensor. Therefore, the effect of temperature on CL signal strength for hairpin structure (HP1 and HP2) incubation was investigated in the range 4–45°C. As shown in Figure 3a, within this range, 37°C was the temperature at which the system generated the best value of CL signal. Because the hairpin structure did not get enough chance to collide at low temperatures, they did not achieve sufficient reaction. In contrast, the excessively high temperature would cause the hairpin structure to be in an unstable state, which would also lead to the decrease of CL signal.

For this system, H_2O_2 is an important factor affecting the CL signal intensity. The concentration and volume of H_2O_2 were optimized in the experiment. As shown in Figure 3b, in the range 10–50 mM, as the concentration of H_2O_2 increases, the intensity of CL signal also increases, and the intensity of CL signal increased to the maximum at 40 mM. In addition, in Figure 3c, the CL signal intensity changes synchronously with the increase of the volume of H_2O_2 involved in the reaction. When the volume of H_2O_2 involved in the reaction was 400 μl, the CL signal intensity increased to a new height. Moreover, when more volumes of H_2O_2 were added, the CL signal did not show any significant change. Therefore, in the following experiment, H_2O_2 with a concentration of 40 mM and a volume of 400 μl was selected to react with the supernatant after magnetic separation to generate the CL signal.

After determining the optimal concentration and volume of the oxide, the injection method selected for the experiment was optimized. The different injection methods have a non-negligible effect on the experimental system, so the optimized injection method is proposed. The experimental results are shown in the Figure 3d. When H_2O_2 was injected into LumAuNPs, the CL intensity is relatively low. In the field of biosensor analysis, the larger the response signal, the higher the sensitivity of the biosensor, so this system was chosen as the injection method for injecting LumAuNPs into H_2O_2.

3.5 | Analytical performance of kanamycin CL detection biosensor

To evaluate the analytical performance of the biosensor, the intensity of the CL signal at different concentrations of kanamycin was measured under the optimal conditions. It can be seen in Figure 4 that the change of CL signal intensity was related to the change of kanamycin concentration, that is, when the kanamycin concentration increased,

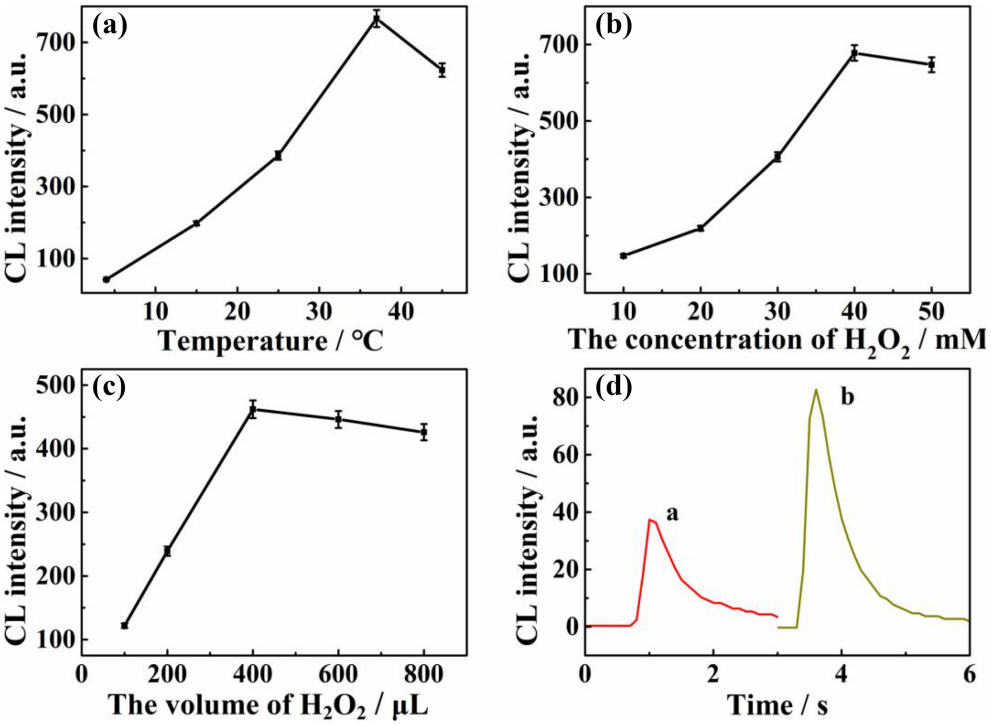


FIGURE 3 (a) Chemiluminescence (CL) intensity produced using different incubation temperatures of HP1 and HP2. Temperature: 4°C, 15°C, 25°C, 37°C, 45°C. (b) CL intensity produced using different concentrations of H_2O_2. Concentrations: 10 mm, 20 mm, 30 mm, 40 mm, 50 mm. (c) CL intensity produced using different volumes of H_2O_2. Volumes: 100 μl, 200 μl, 400 μl, 600 μl, 800 μl. (d) CL intensity produced using different implantation methods: injecting H_2O_2 into LumAuNPs (a) and injecting LumAuNPs into H_2O_2 (b)

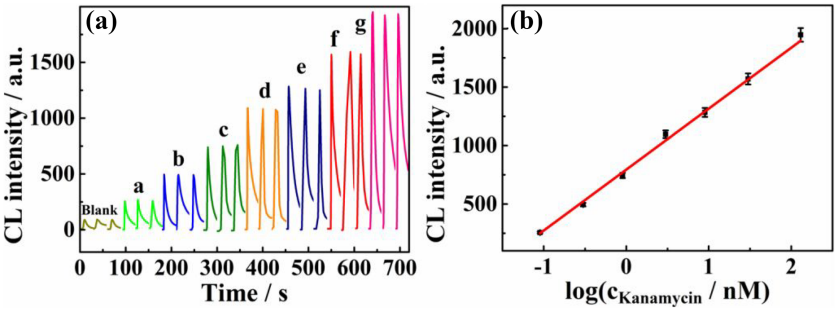


FIGURE 4 (a) Chemiluminescence (CL) intensity of kanamycin at concentrations of 0.09 nM (sample a), 0.3 nM (sample b), 0.9 nM (sample c), 3 nM (sample d), 9 nM (sample e), 30 nM (sample f), and 130 nM (sample g). (b) Linear relationship between CL intensity and kanamycin concentration.

TABLE 1 Comparison of different methods for kanamycin assay

Method[a]	Signal source[b]	Linear range[c]	LOD[c]	Ref.
Col	AuNPs	5.82–23303.2	4.66	[25]
Flu	FAM-cDNA	0.58–29.13	0.23	[26]
Flu	Rhodamine B	14.42–79.90	4.37	[27]
PEC	Methylene blue	14.56–524.32	7.57	[28]
CL	LumAuNPs–sDNA	0.052–75.74	0.023	This work

[a]Col, colorimetry; Flu, fluorescence; PEC, photoelectrochemistry.
[b]Signal source in different systems.
[c]μg/kg.

the CL signal intensity also increased. With the kanamycin concentrations ranging from 0.09 to 130 nM, a good linear relationship was shown between CL signal intensity and kanamycin concentration, and the regression equation was I = 521.252logC+749.399 and a correlation coefficient of 0.9962. The detection limit was 0.04 nM, and the detection limit of the system without adding HP2 was 0.22 nM. The comparison of the detection limit shows that the signal amplification factor of the system was 5.5 times. Table 1 shows the comparison of the performance of different analytical methods in detecting kanamycin. This proved that the constructed biosensor for kanamycin detection using the CL method was very efficient and accurate, and the experimental results were reliable.

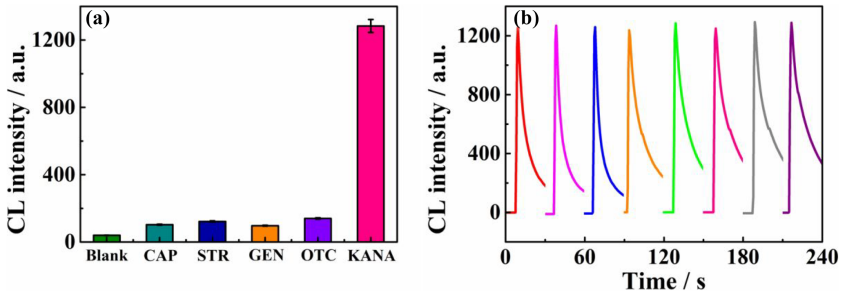


FIGURE 5 (a) Chemiluminescence (CL) reaction intensity of different antibiotics in this system. (b) Intensity of CL signal with 20 nM kanamycin based on eight repeated detection cycles

TABLE 2 Precision (RSD, %) and accuracy (recovery, %) of kanamycin added to the fish samples (nM, $n = 6$)

Original	Added	Found		RSD	Recovery
		This method	Official method[a]		
–[b]	0.10	0.101	0.096	4.8	101.0
–	10.0	9.723	9.781	5.2	97.2
–	100	98.663	99.012	5.4	98.7

[a]Liquid chromatography–tandem mass spectrometry.
[b]Kanamycin concentration below LOD.

3.6 | Specificity and reproducibility of kanamycin CL biosensor

Specificity is an important parameter for biosensor performance, so kanamycin (KANA) and four similar antibiotics: streptomycin (STR), chloramphenicol (CAP), gentamicin (GEN) and oxytetracycline (OTC), were used to test biosensor specificity. As shown in Figure 5a, a weak CL signal was generated in the absence of kanamycin. Only when kanamycin was present, the system generated a strong CL signal. This showed that the constructed biosensor has excellent specificity for kanamycin.

Repeatability is important for biosensor applications, the relative standard deviation (RSD) was chosen to measure the stability of the biosensor. The results of eight repeated detection cycles of the biosensor with a concentration of 20 nM kanamycin are shown in Figure 5b. According to the peak value of CL system, the CL intensity was quite stable with an RSD of 1.8%, which is sufficient to indicate excellent reproducibility of the biosensor.

3.7 | Real sample detection

To evaluate the feasibility of the kanamycin CL biosensor for the detection of kanamycin residue levels in actual samples, the standard addition method was used to detect the accuracy of kanamycin, and the standard addition method was used to calculate the kanamycin recovery rate. As shown in Table 2, the recovery rate of kanamycin with a concentration ranging from 0.1 nM to 100 nM is between 97.2% and 101%. Compared with the official method, this method has good accuracy in the detection of kanamycin in fish samples.

4 | CONCLUSION

In summary, we designed an analytical strategy using LumAuNPs as probe material, combined with MBs and catalytic hairpin assembly technology, which realized the recycling of kanamycin molecule, amplified CL signal, and detected kanamycin residue level in fish by CL analysis. Compared with the existing traditional methods, this strategy has many advantages. Firstly, LumAuNPs with good CL performance and catalytic function has been successfully used as the probe material for kanamycin detection, which is of great significance for the application of nanomaterials in the field detection of antibiotics. Secondly, the system has specificity and designability. The application of high-quality aptamer technology in the detection of kanamycin in the concentration range 0.09–130 nM also has good practicability compared with the standard method, which provides a basis for the rapid and simple detection of kanamycin in samples in the future, and can also be used for specific detection of other antibiotics through correct DNA design. In short, this method is a simple and reliable means of detecting kanamycin, and it will have great development prospects in the field of antibiotic detection in the future.

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SUPPORTING INFORMATION

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