

Investigation on Ruthenium(II) Bipyridine/Ag^{III} Complexes Chemiluminescence System and Its Application for Sensitive Norfloxacin and Ofloxacin Detection

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The reaction mechanism of Ag^{III} complex ([Ag(HIO₆)₂]⁵⁻) with ruthenium(II) bipyridine (Ru(bpy)₃²⁺) in acid medium was investigated. A novel flow injection chemiluminescence (CL) analysis method was developed for the detection of norfloxacin (NFLX) and ofloxacin (OFLX) in commercial drug, milk and human urine samples based on Ag^{III}-ruthenium(II) bipyridine in acidic solution. Under optimal conditions, CL intensities were proportional to drug concentrations in the range of 1.6×10^{-9} to 8×10^{-6} g mL⁻¹ for NFLX and 1.0×10^{-9} to 4×10^{-6} g mL⁻¹ for OFLX. The detection limit was 1.0×10^{-9} g mL⁻¹ for NFLX and 0.8×10^{-9} g mL⁻¹ for OFLX. On the basis of the CL, UV and fluorescence spectra, possible reaction mechanisms were proposed.

Keywords: Ag^{III}, ruthenium(II) bipyridine, CL, mechanism

Introduction

Norfloxacin (NFLX) and ofloxacin (OFLX) belong to the third generation of quinolone antibiotics with a broad spectrum of activity widely used in human and veterinary medicine. NFLX and OFLX inhibit the activity of DNA helicase and topoisomerase IV in digestive tract pathogenic bacteria, achieving rapid bactericidal effects. The abuse of quinolones in animal husbandry has led to serious food safety problems.¹ It is, therefore, necessary to rapidly and sensitively detect fluoroquinolones in biological samples and foods.

Flow-injection chemiluminescence (CL) methods have received much attention for analytical purposes due to their advantages of low cost, excellent sensitivity and rapid analysis. Several CL methods within various reaction systems have been used for the analysis of NFLX and OFLX, including Ru(bpy)₃²⁺-Ce^{IV}-H₂SO₄,² cerium(IV)-sulfite-terbium(III),³ hydrogen peroxide-sulfuric acid,⁴ luminol-hydrogen peroxide-nano gold,^{5,6} Ce^{IV}-Na₂SO₃,⁷ Ce^{IV}-Na₂S₂O₄,^{8,9} and Ag^{III}-H₂SO₄.^{10,11} The detection limit of these methods ranges from 1.1×10^{-10} to 2.61×10^{-8} g mL⁻¹. The dynamic linear range can be as low as 10 times, and as high as 400 times.

In our previous studies,^{12,13} we have been focusing on the interactions between the bis(hydrogenperiodato) argentate(III) complex anion, [Ag(HIO₆)₂]⁵⁻, and bioactive and medically important molecules in alkaline medium. Based on the Ag-luminol-NaOH CL system, a variety of sensitive detection methods for small biological molecules¹⁴⁻¹⁶ and drug molecules^{17,18} have been developed. However, the oxidation properties and applications of Ag^{III} in acidic medium require further analysis. We found that the oxidizing ability of Ag^{III} in acidic medium was stronger than that in alkaline medium. Ag^{III} in acidic medium can not only oxidize fluoroquinolones, but can also oxidize water to oxygen,¹¹ and oxidize ruthenium(II) to ruthenium(III). As a result, the chemical reaction and luminescence mechanism of Ag^{III}-Ru^{II}-sulfuric acid system was studied in detail using UV, fluorescence and CL spectra. Due to fluoroquinolones sensitizing this system, a new method for the determination of NFLX and OFLX in pharmaceutical and biological samples based on the Ag^{III}-Ru^{II}-sulfuric acid system was proposed.

Experimental

Chemicals and solutions

All chemicals were of analytical reagent grade and

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ultrapure water was used throughout. The Ag^{III} complex was synthesized according to previously described procedures.¹⁴ The concentration of stock solution was determined according to the molar absorptivity ($\epsilon = 1.26 \times 10^4 \text{ mol}^{-1} \text{ L cm}^{-1}$) at 362 nm. Solutions were stored at room temperature and diluted to working concentrations in pure water.

The Ru(bpy)₃Cl₂·6H₂O (tris(2,2'-bipyridine) ruthenium complex) was purchased from Sigma-Aldrich. Ru(bpy)₃Cl₂·6H₂O was dissolved in pure water to a stock concentration of $4 \times 10^{-3} \text{ mol L}^{-1}$. Stock solutions were stored at 4 °C and diluted to working solutions in water.

Stock solutions of 1 mg L⁻¹ OFLX and NLFX (Institute for the Control of Pharmaceutical and Biological Products) were prepared by dissolving 10 mg in 0.5 mL 0.1 mol L⁻¹ hydrochloric acid and diluting to 10 mL with pure water.

Instrumentation

The flow-injection analysis (FIA) CL system (Xi'an Remax Electronic Science-Tech Co. Ltd., shown in Figure 1) consisted of two peristaltic pumps, a six-way injection valve and photomultiplier tube detector. [Ag(HIO₆)₂]⁵⁻ and Ru(bpy)₃²⁺ solution were delivered by pump P1. Solutions were mixed and produced a baseline signal. Sample solutions were injected in a six-way injection valve with a sample loop by P2. Sample solution, which was carried by the [Ag(HIO₆)₂]⁵⁻ stream, were mixed with acidic ruthenium(II) bipyridine solution to produce enhanced CL signals. CL was detected through a photomultiplier tube detector (PMT, operated at -600 V).

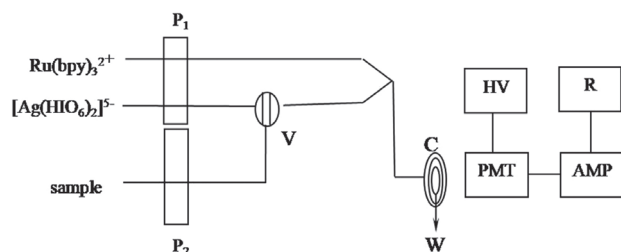


Figure 1. Schematic diagram of flow injection CL analysis system. P, peristaltic pump; V, sampling inlet valve; C, flowing cell; PMT, photomultiplier tube; AMP, amplifier; HV, high voltage; R, recorder; W, waste.

UV absorbance was assessed using TU-1901 UV-Vis spectrophotometry (Beijing Purkinje General Instrument Co. Ltd.). Fluorescence spectra were obtained using an F-7000 fluorescence spectrophotometer (Hitachi). CL spectral information was achieved using 20 narrow band interference filters (360-670 nm), which were inserted between the cuvette and photomultiplier tube.

Sample treatment

Drug capsules

Ten capsules of the same batch number (drug labeled quantity 0.1 g *per* granules) were randomly selected and taken as the total mass. It was dissolved 1/10 of the total mass (equivalent to the mass of a single capsule) in a small volume of 0.1 mol L⁻¹ hydrochloric acid and diluted the sample in water to the required concentration.

Milk

Milk samples (1 mL) purchased from a local market were diluted to 10 mL in water. From diluted samples, 4 mL was added to 1 mL of 20% trichloroacetic acid, and centrifuged at 14000 rpm for 10 min. Supernatants were collected and analyzed.

Urine samples

Urine was added to 0.2 g PbO₂ to remove uric acid, thiourea, and ascorbic acid. Samples were vortexed for 5 min and centrifuged at 4000 rpm for 10 min. Supernatants were collected, 1 mL of which was diluted to a final volume of 10 mL in distilled water for analysis.

Results and Discussion

Kinetics of the CL reaction

Investigation of the CL reaction kinetic curves was performed using the static CL analysis method. Briefly, in a 10-mL beaker, 0.2 mL of fluoroquinolones and 0.2 mL H₂SO₄ (0.2 mol L⁻¹) were mixed, and 0.2 mL [Ag(HIO₆)₂]⁵⁻ was injected into the beaker. A small CL signal (Figure 2, curve a) was observed. When 0.2 mol L⁻¹ H₂SO₄ was replaced with 2 μmol L⁻¹ Ru(bpy)₃²⁺ solution (containing 0.2 mol L⁻¹ H₂SO₄), and [Ag(HIO₆)₂]⁵⁻ re-injected, CL signals became significantly enhanced (Figure 2, curve b). The reaction rate in the [Ag(HIO₆)₂]⁵⁻-Ru(bpy)₃²⁺-H₂SO₄-NFLX system was rapid, with only 0.2 s required from reagent mixing to peak maximum signals. The reaction signal returned to zero after 6 s.

UV spectra

UV spectra for different systems (Figures 3 and 4) were examined to obtain further information regarding the CL mechanism of the [Ag(HIO₆)₂]⁵⁻-H₂SO₄-fluoroquinolones-Ru(bpy)₃²⁺ system. Figure 3 shows that Ru^{II} displayed absorption bands at approximately 288 nm (curve a). Ag^{III} possesses a characteristic absorption band at 357 nm (curve b). When Ag^{III} was mixed with acid, the absorption

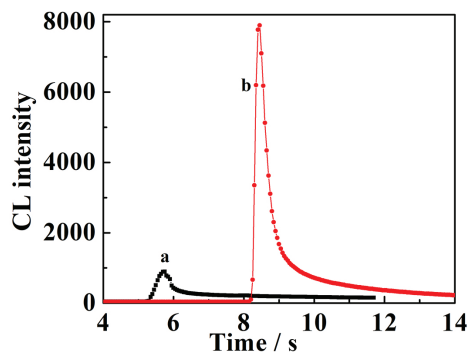


Figure 2. Kinetic characteristic of the CL reaction. (a) $[\text{Ag}(\text{HIO}_6)_2]^{5-}$ - H_2SO_4 - Ru^{II} system; (b) $[\text{Ag}(\text{HIO}_6)_2]^{5-}$ - H_2SO_4 - Ru^{II} -OFLX system. $[\text{Ag}(\text{HIO}_6)_2]^{5-}$, 1.0×10^{-4} mol L^{-1} ; OFLX, 1×10^{-6} g mL^{-1} ; H_2SO_4 , 0.2 mol L^{-1} . PMT, 800 V.

peak of $[\text{Ag}(\text{HIO}_6)_2]^{5-}$ almost disappeared (curve d). The curve c was the sum of curves a and b, which showed that Ru^{II} was minimally oxidized by Ag^{III} . When sulfuric acid was added to the system, peaks characteristic of Ru^{II} and Ag^{III} disappeared (curve e), indicating that Ru^{II} is easily oxidized by Ag^{III} in acid medium.

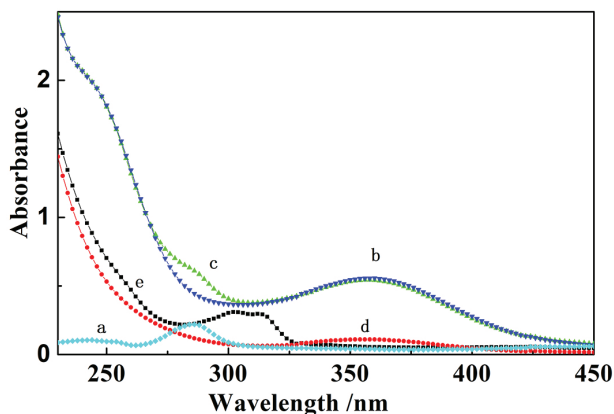


Figure 3. UV spectra of Ag^{III} - Ru^{II} - H_2SO_4 systems. (a) $\text{Ru}(\text{bpy})_3^{2+}$; (b) $[\text{Ag}(\text{HIO}_6)_2]^{5-}$; (c) $[\text{Ag}(\text{HIO}_6)_2]^{5-}$ - $\text{Ru}(\text{bpy})_3^{2+}$; (d) $[\text{Ag}(\text{HIO}_6)_2]^{5-}$ - H_2SO_4 ; (e) $[\text{Ag}(\text{HIO}_6)_2]^{5-}$ - $\text{Ru}(\text{bpy})_3^{2+}$ - H_2SO_4 . $\text{Ru}(\text{bpy})_3^{2+}$, 5×10^{-6} mol L^{-1} ; $[\text{Ag}(\text{HIO}_6)_2]^{5-}$, 2.5×10^{-5} mol L^{-1} ; H_2SO_4 , 0.2 mol L^{-1} .

Figure 4 indicates that NFLX alone has a maximum absorption peak at a wavelength of approximately 277 nm (curve a). Following mixing of NFLX with Ag^{III} , the $[\text{Ag}(\text{HIO}_6)_2]^{5-}$ color slowly faded and the absorption peak of Ag^{III} decreases gradually (curve c). However, when acid and NFLX were mixed with Ag^{III} , the absorption peak and color of Ag^{III} disappeared rapidly (curve d). Figures 3 and 4 display the oxidative ability of the Ag^{III} complex is improved through the addition of acidic medium as a catalyst.

Fluorescence spectra

Fluorescence emission spectra of NFLX and OFLX were observed in Figures 5A and 5B. As shown in

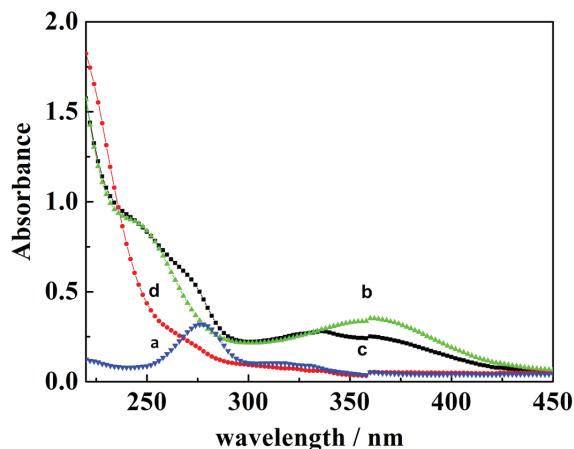


Figure 4. UV spectra of Ag^{III} -NFLX- H_2SO_4 system. (a) NFLX; (b) $[\text{Ag}(\text{HIO}_6)_2]^{5-}$; (c) $[\text{Ag}(\text{HIO}_6)_2]^{5-}$ -NFLX; (d) $[\text{Ag}(\text{HIO}_6)_2]^{5-}$ -NFLX- H_2SO_4 . NFLX, 1.25×10^{-6} g mL^{-1} ; $[\text{Ag}(\text{HIO}_6)_2]^{5-}$, 2.0×10^{-5} mol L^{-1} ; H_2SO_4 , 0.2 mol L^{-1} .

Figure 5A, acidic NFLX produced fluorescence emission at 475 nm (curve a). $\text{Ru}(\text{bpy})_3^{2+}$ produced fluorescence emission at 590 nm (curve b). When $[\text{Ag}(\text{HIO}_6)_2]^{5-}$ was added to the acidic $\text{Ru}(\text{bpy})_3^{2+}$ and NFLX mixed solution,

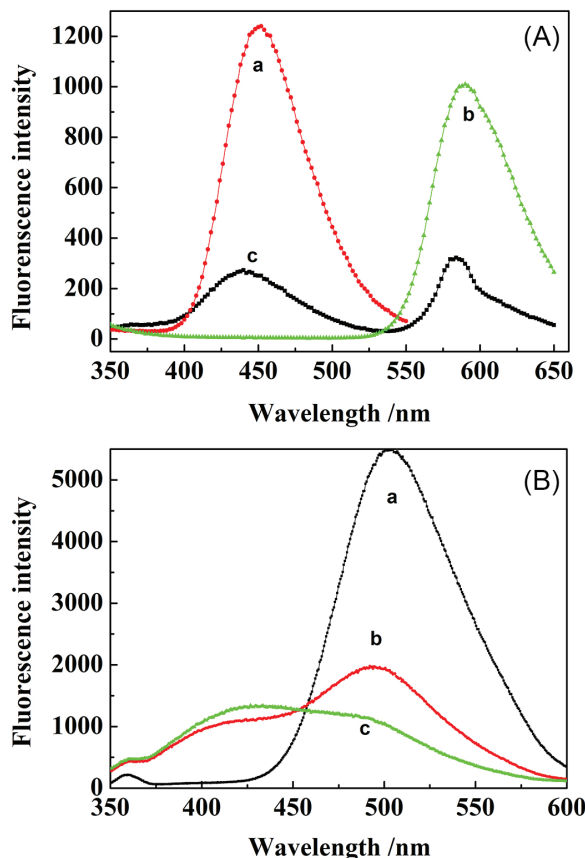


Figure 5. Fluorescence spectra of different systems. (A) a, acidic NFLX; b, $\text{Ru}(\text{bpy})_3^{2+}$; c, acidic $\text{Ru}(\text{bpy})_3^{2+}$ + NFLX + $[\text{Ag}(\text{HIO}_6)_2]^{5-}$; $\lambda_{\text{ex}} = 290$ nm; (B) a, acidic OFLX; b and c, acidic $\text{Ru}(\text{bpy})_3^{2+}$ + OFLX + $[\text{Ag}(\text{HIO}_6)_2]^{5-}$; $\lambda_{\text{ex}} = 320$ nm. NFLX, 1.0×10^{-6} g mL^{-1} ; OFLX, 1.0×10^{-6} g mL^{-1} ; $\text{Ru}(\text{bpy})_3^{2+}$, 6×10^{-6} mol L^{-1} ; $[\text{Ag}(\text{HIO}_6)_2]^{5-}$, 1×10^{-4} mol L^{-1} ; H_2SO_4 , 0.2 mol L^{-1} .

the fluorescence peaks at 475 and 590 nm decreased rapidly (curve c). Figure 5B shows that the acidic OFLX produced fluorescence emission at 505 nm (curve a). When [Ag(HIO₆)₂]⁵⁻ was added into acidic OFLX, the fluorescence peak at 505 nm decreased gradually, but the fluorescence peak at approximately 430 nm gradually increased (curves b and c). This demonstrates that OFLX is oxidized to form a fluorescent product near 430 nm.

CL spectra

The mechanism of the CL reaction was investigated using the CL spectra (Figure 6). From Figure 6 (curves a and b) it can be seen that the maximal CL spectrum was located at about 610 nm, the characteristic wavelength of Ru(bpy)₃^{2+*}.¹⁹ In addition, relatively weak CL emission at 490 and 430 nm was also observed in the two CL systems. According to previous study,¹¹ the [Ag(HIO₆)₂]⁵⁻-H₂SO₄ system produces CL emission at 490 nm, which may have been caused by the excited state (O₂)₂. CL emission at 440 nm (Figure 6, curve b) may have occurred due to OFLX intermediate product energy transfer, the fluorescence peak of which is 430 nm (Figure 5, curve c).

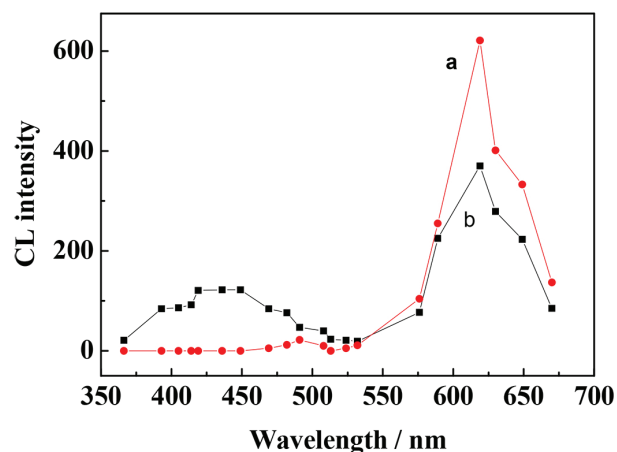


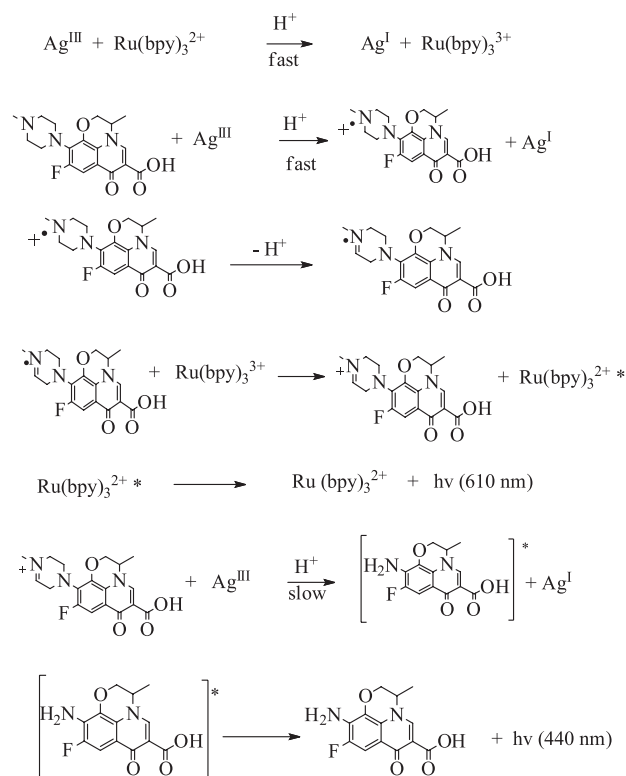
Figure 6. CL spectra. (a) [Ag(HIO₆)₂]⁵⁻-Ru(bpy)₃²⁺-NFLX-H₂SO₄ system; (b) [Ag(HIO₆)₂]⁵⁻-Ru(bpy)₃²⁺-OFLX-H₂SO₄ system. OFLX, 1.0 × 10⁻⁶ g mL⁻¹; NFLX, 1.0 × 10⁻⁶ g mL⁻¹; Ru(bpy)₃²⁺, 2 × 10⁻⁶ mol L⁻¹; [Ag(HIO₆)₂]⁵⁻, 1 × 10⁻⁴ mol L⁻¹; H₂SO₄, 0.4 mol L⁻¹; PMT, 800 V.

Possible CL reaction mechanism

To study whether Ru(bpy)₃³⁺ can be directly reduced to Ru(bpy)₃^{2+*} by fluoroquinolones, it was prepared Ru(bpy)₃³⁺ using PbO₂ and observed the CL spectra of the Ru(bpy)₃³⁺-fluoroquinolones-H₂SO₄ system.²⁰ The CL intensity was too small to observe, indicating that the fluoroquinolones reactive intermediates should be reacted with Ru(bpy)₃³⁺ rather than the actual original form of fluoroquinolones.

Both UV absorption and fluorescence spectra suggested that chemical reactions had occurred between [Ag(HIO₆)₂]⁵⁻ and Ru(bpy)₃²⁺ in addition to [Ag(HIO₆)₂]⁵⁻ and fluoroquinolones in H₂SO₄ medium. The mechanism of luminescence can be summarized as follows: ruthenium(II) is first oxidized to ruthenium(III) by Ag^{III}; then, ruthenium(III) is reduced to excited ruthenium(II) by the fluoroquinolones active intermediates, which are produced through the reaction of Ag^{III} and fluoroquinolones; and finally, excited ruthenium(II) emits characteristic spectra at about 610 nm and meanwhile become ground state ruthenium(II).

Based on our experiments and previous studies,^{21,22} taking OFLX as an example, a specific CL mechanism is described in Scheme 1.



Scheme 1. Possible mechanism for the OFLX-Ag^{III}-Ru(bpy)₃²⁺-H₂SO₄ CL system.

Optimization of experimental conditions

Experimental conditions were optimized through orthogonal experiments using three factors and three levels. Ag^{III} as the oxidant, H₂SO₄ as the acid medium and Ru(bpy)₃²⁺ as the CL reagent were set as the three factors for orthogonal experiments. The experimental results show that Ru(bpy)₃²⁺ had the largest CL intensity influence factor, the second being the acid concentration, and the third being the Ag^{III} concentration. Conditions for subsequent experiments were selected as 3 × 10⁻⁶ mol L⁻¹ Ru(bpy)₃²⁺,

$1.0 \times 10^{-4} \text{ mol L}^{-1} \text{ Ag}^{\text{III}}$, $0.3 \text{ mol L}^{-1} \text{ H}_2\text{SO}_4$ for OFLX and 0.2 mol L^{-1} for NFLX.

Analytical characteristics

Under the optimal conditions described, the calibration graphs of NFLX were linear in the range of 1.6×10^{-9} - $8 \times 10^{-6} \text{ g mL}^{-1}$. The equation of linear regression was $I = 2779 \times 10^6 C + 66.22$ (regression coefficient (r) = 0.9984). The detection limit of NFLX ($S/N = 3$) was $1 \times 10^{-9} \text{ g mL}^{-1}$ and relative standard derivation (RSD) was 1.34% for $1 \mu\text{g mL}^{-1}$ NFLX ($n = 11$). Within 180 min, the 7 measurements of CL intensity had a variation of 3.06%.

The calibration graph of OFLX was linear in the range of 1×10^{-9} - $4 \times 10^{-6} \text{ g mL}^{-1}$. The equation of linear regression was $I = 965.92 \times 10^6 C + 23.875$ ($r = 0.9991$), and the detection limit was $0.8 \times 10^{-9} \text{ g mL}^{-1}$. Relative standard derivation for OFLX ($n = 7$) was 1.10%. Within 180 min, the 7 measurements of CL intensity had a variation of 1.65%. Both the detection limit and linear range of our assays were performed to a higher level than previous methods (Table 1). Particularly for the dynamic linear range, the present study displayed nearly 5000 times. Such a wide dynamic linear range is rare in CL analysis.

The effects of common substances in drug and urine sample on the determination of $1 \mu\text{g mL}^{-1}$ OFLX and NFLX were examined. A foreign substance is considered not to interfere such as $100 \mu\text{g mL}^{-1} \text{ Na}_2\text{SO}_4$, CaSO_4 , KNO_3 ,

ZnSO_4 and polyglycol, $68 \mu\text{g mL}^{-1}$ uric acid, $10 \mu\text{g mL}^{-1}$ glucose and lactose, $4 \mu\text{g mL}^{-1}$ glutathione due to relative error were $\leq \pm 5\%$.

Sample analysis

The proposed method was applied to determine the content of NFLX and OFLX in capsules and tablets. The results and recoveries are shown in Table 2 along with the results obtained using the UV method, which were in excellent agreement.

To evaluate the validity of the proposed method for the determination of milk and urine sample, recovery studies were performed using matrix labeling working curves. The recovery values of milk for NFLX were in the range of 98.51-105.17% with RSDs of 3.17-3.58%.

Urine samples were taken from two healthy volunteers. NFLX and OFLX (200 mg) were taken orally by the subjects and assessed after 2 h. Results are presented in Table 3. The recovery values for NFLX and OFLX were in the range of 97.2-105.6% with RSDs of 0.35-3.58%.

Conclusions

This work demonstrates a new CL system composed of Ag^{III} complexes as oxidant and acidic ruthenium(II) as a CL reagent. Fluoroquinolones drugs could dramatically enhance CL intensity in this system. Thus, accurate and sensitive detection of OFLX and NFLX in drug, milk and

Table 1. Comparison of various CL systems for the determination of fluoroquinolones

Method		Linear range	Detection limit / (g mL^{-1})	Reference
$\text{Ru}(\text{bpy})_3^{2+}\text{-Ce}^{\text{IV}}$	ofloxacin	$0.003\text{-}0.7 \mu\text{g mL}^{-1}$	1.1×10^{-10}	2
	norfloxacin	$0.05\text{-}7.0 \mu\text{g mL}^{-1}$	1.75×10^{-9}	
$\text{Ce}^{\text{IV}}\text{-Na}_2\text{SO}_3$	ofloxacin	$0.04\text{-}4 \mu\text{g mL}^{-1}$	1.6×10^{-8}	7
$\text{Ce}^{\text{IV}}\text{-Na}_2\text{S}_2\text{O}_4$	ofloxacin	$2.0 \times 10^{-8}\text{-}6.0 \times 10^{-6} \text{ g mL}^{-1}$	7×10^{-9}	8
	norfloxacin	$1.0 \times 10^{-8}\text{-}6.0 \times 10^{-5} \text{ g mL}^{-1}$	6×10^{-9}	
$\text{Ce}^{\text{IV}}\text{-Na}_2\text{SO}_3\text{-nano Au}$	norfloxacin	$7.9 \times 10^{-7}\text{-}1.9 \times 10^{-5} \text{ g mL}^{-1}$	2.61×10^{-8}	5
$\text{Ag}^{\text{III}}\text{-H}_2\text{SO}_4$	norfloxacin	$1.34 \times 10^{-8}\text{-}5.44 \times 10^{-6} \text{ g mL}^{-1}$	3.10×10^{-9}	10
$\text{Ru}(\text{bpy})_3^{2+}\text{-Ag}^{\text{III}}\text{-H}_2\text{SO}_4$	ofloxacin	$1.0 \times 10^{-9}\text{-}4 \times 10^{-6} \text{ g mL}^{-1}$	0.8×10^{-9}	this method
	norfloxacin	$1.6 \times 10^{-9}\text{-}8 \times 10^{-6} \text{ g mL}^{-1}$	1×10^{-9}	

Table 2. Analytical results of ofloxacin (OFLX) and norfloxacin (NFLX) in drug samples

Analyte	Labeled value / (mg per capsule)	Determination value		Added / mg	Found / mg	Recovery / %	RSD ($n = 3$) / %
		This method	UV				
Norfloxacin	100	97.0	99.2	50	51.1	102.6	1.5
				100	99.9	97.4	1.04
				200	206	109.5	1.12
Ofloxacin	100	91.0	92.2	50	52.8	105.6	0.96
				100	102.4	102.4	0.39
				200	198	99.0	0.87

RSD: relative standard deviation.

Table 3. Determination results of ofloxacin (OFLX) and norfloxacin (NFLX) in milk and urine sample

Sample/analyte	Background	Added / ($\mu\text{g mL}^{-1}$)	Found / ($\mu\text{g mL}^{-1}$)	Recovery / %	RSD (n = 3) / %
Milk/norfloxacin	0	0.1	0.096	98.51	3.53
		0.2	0.202	100.89	3.58
		0.5	0.526	105.17	3.17
Urine/norfloxacin	6.85	2.5	9.28	97.2	0.72
		5.0	12.04	103.8	0.59
		10.0	16.83	99.98	0.55
Urine/ofloxacin	4.52	2.5	7.13	104.4	0.35
		5.0	9.80	105.6	0.46
		10.0	14.89	103.7	0.53

RSD: relative standard deviation.

human urine samples was achieved. An advantage of this method was that the dynamic linear range was up to 3–4 orders of magnitude and the detection limit was as low as $0.8 \times 10^{-9} \text{ g mL}^{-1}$. The possible CL enhance mechanism of the system was deeply researched.

The UV spectra, fluorescence spectra and CL spectra show that the Ag^{III} complexes exhibit strong oxidation ability in acidic media, which not only increases the rate of drug oxidation, but also oxidizes water to oxygen and ruthenium(II) to ruthenium(III). It can be concluded that the Ag^{III} complexes are similar to strong oxidants potassium permanganate, that is, they possess oxidation ability in both acidic and alkaline media, but display stronger oxidative ability in acidic media.

Our work broadens the range of applications of Ag^{III} reagents and species of chemiluminescence systems.

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