Talanta 88 (2012) 168–174

Contents lists available at SciVerse ScienceDirect

Talanta

journal homepage: www.elsevier.com/locate/talanta

Simultaneous laser-induced fluorescence, coaxial thermal lens spectroscopy and retro-reflected beam interference detection for capillary electrophoresis

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ARTICLE INFO

Article history:
Received 6 August 2011
Received in revised form 18 October 2011
Accepted 22 October 2011
Available online 25 October 2011

Keywords:
Capillary electrophoresis
Coaxial thermal lens spectroscopy
Laser-based fluorescence
Retro-reflected beam interference
Simultaneous detection

ABSTRACT

A simultaneous laser-induced fluorescence (LIF), coaxial thermal lens spectroscopy (TLS) and retro-reflected beam interference (RBI) detection for capillary electrophoresis (CE), has been described. In its optical scheme, a diode-pump solid-state (DPSS) laser was employed as the pump laser in both LIF detection and coaxial TLS detection, and a He–Ne laser was utilized as the probe laser in coaxial TLS detection and RBI detection. In addition, RBI signals with and without thermal lens had been theoretically compared to ensure the reliability of the RBI signal. Moreover, the focus length of the key lens has been optimized to improve the performance of the proposed detection. The last but not least, several determinations were taken to evaluate its limit of detection, linear range as well as relative standard deviation, all of which indicate no worse results compared with former reports. LIF and coaxial TLS detection owned high sensitivity, and RBI detection indicated versatile property, based on which the reported detection achieved a sensitive and universal detection for CE.

1. Introduction

A prominent area in CE researches is the pursuit of a sensitive detection with the versatile property. Unfortunately, for most CE detections, there are certain limitations [1,2]. Because of the background current in electrophoresis and the absence of electrochemical activity for most chemicals [3,4], electrochemical CE detections were not so satisfactory. The daily maintenance of CE–MS is still troublesome [5,6]. Most unconventional detections, such as NMR detection and radioisotope detection performed poorly in either quantitative applications or sensitivity [7–10]. While UV–vis absorbance detection was a common, straightforward means to realize on-column detection, achievable sensitivity is limited for CE [11–13]. LIF detection shows obvious advantages in sensitivity, while it is usually accompanied with the derivatization, which is troublesome and burdensome [14]. Although indicating high sensitivities, chemiluminescence detection [15–17] and TLS detection [18–20] are still confined to some analytes. Refractive index (RI) detection, whereas universal and quantitative, has not been comprehensively applied because of its unsatisfactory sensitivity and complicated optical schemes [21–27]. Compared with other detectors, optical detectors indicated advantages in sensitivity, on-column detection, daily maintenance, cost and realization [28]. In this sense, it will be significant to develop a sensitive and versatile optical detection for CE.

Since there is no one among preceding laser-based detections achieving a sensitive and versatile detection, some publications tried to seek their combinations. Bornhop and Dovichi described a cross-beam TLS detection coupled with RI detection for static samples in capillary, which was based on a complicated optical scheme [29]. A combination of absorbance and RI detection, which was based on Fabry-Perot interferometry, was reported by Yeung and coworkers [30]. In addition, they described another combination of absorbance, fluorescence and RI detection [31]. Then, Abbas and Shelly introduced another simultaneous absorbance, fluorescence and RI detection for liquid chromatography [32]. It was worthy to emphasize that preceding three detections were for liquid chromatography and based on a designed cell. Moreover, neither of preceding strategies included the tightly focusing process for the laser beam, which played a vital role in both LIF and TLS detection. Generally, a sensitive and versatile laser-based detection for CE is still absent.

Abbreviations: LIF, laser-induced fluorescence; TLS, thermal lens spectroscopy; RBI, retro-reflected beam interference; CE, capillary electrophoresis; DPSS, diode-pump solid-state; RI, refractive index; LOD, limit of detection; RSD, relative standard deviation.

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doi:10.1016/j.talanta.2011.10.027

Some years later, Kitamorie and coworkers reported a simultaneous LIF and TLS detection [33], while both detections indicated poor versatile property. On the other hand, Deng and Li reported the retro-reflected beam interference (RBI) technique to improve the RI detection for CE [34]. A tightly focusing process for excitation laser beam, which was indispensable in LIF and TLS detection, was included in RBI optical scheme. Moreover, RBI detection is feasible to be integrated with TLS and LIF detection, respectively [35,36]. In this case, it is spontaneous to seek a simultaneous LIF, coaxial TLS and RBI detections for CE, which should be sensitive and universal. To the best of our knowledge, the proposed research should be the first report for simultaneous LIF, coaxial TLS and RBI detection for CE.

In this paper, a simultaneous LIF, coaxial TLS and RBI detection for CE has been described. In addition, RBI signals with and without thermal lens had been theoretically compared to ensure a reliable RBI signal. Furthermore, the lens focus in the reported detection had been optimized to improve its performance. At last but not least, some samples were determined to evaluate its limit of detection (LOD), linear range and relative standard deviation (RSD) for the proposed detection, as well as to indicate its advantage in the combination of high sensitivity and versatile property.

2. Materials and methods

2.1. Optical scheme and signal modulation

The optical scheme of simultaneous LIF, coaxial TLS and RBI detection has been indicated in Fig. 1(a). A DPSS laser Compass 215M-20 (Coherent, CA, USA) produced a linearly polarized 20 mW pump laser beam at 532 nm with a polarizer. A quarter-wave retardation plate was utilized behind the polarizer to extinguish retro-reflected light. After reflected by a reflector, the pump beam was modulated by a mechanical chopper, which worked at 300 Hz. Then, the pump beam was focused by a 10.5 mm lens and introduced into a capillary without outer protection coatings. A He–Ne laser Hn-250A (Hongyang, Shanghai, China) produced a linearly polarized 2 mW probe beam at 632.8 nm by a polarizer and a quarter-wave retardation plate. Then, two 50:50 beam splitters at 532 nm were employed. One was used to get the DPSS laser beam and the He–Ne laser beam coaxial, and the other one reflected RBI fringes to the perpendicular direction. After a pinhole and a high-pass filter at 560 nm, initial RBI signal was obtained from a photodiode S4349 (Hamamatsu, Japan). In perpendicular direction, an objective was utilized to collect fluorescence, while scattering light from the capillary had been filtered to improve its signal–noise ratio with a pinhole and a band-pass filter at 560 nm (±10 nm). Then, the fluorescence was monitored as initial LIF signal by a photomultiplier tube PMT1 CR131 (Hamatatsu, Beijing, China). The coaxial beam, after reflected by a reflector, propagated to another photomultiplier tube PMT2 CR131 (Hamatatsu, Beijing, China), which provided initial coaxial TLS signal. All optical components, fixtures, translation stages and adaptors were obtained from Zolix Instruments, Beijing, China.

In the reported scheme, chromatic aberration was employed to mismatch the pump laser waist and the probe beam waist. Two laser beams have been collinear at the first beam splitter, after which the collinear beam was focused into the capillary. Because of their wavelength differences, the mismatch between the pump laser waist and the excitation beam waist had been automatically formed to realize the coaxial TLS detection.

The block diagram of signal modulations in the proposed detection was given in Fig. 1(b). The initial LIF signal and the initial coaxial TLS signal were transmitted to a lock-in amplifier ND-201 (NDWSHB, Nanjing, China), while the chopping frequency from the chopper ND-4 (NDWSHB, Nanjing, China) was set as the reference signal. In addition, initial RBI signal was conditioned by a current-to-voltage signal amplifier, which was composed by a 5 MΩ feedback resistor in parallel with an operation amplifier 129P (Texas Instrument, TX, USA). At last, signals from the lock-in amplifier and the output of the preceding signal amplifier were collected as LIF signal, coaxial TLS signal and RBI signal, respectively.

2.2. CE operation

A 30 kV DC high voltage supply DW-P303-1AC (Dongwen, Tianjin, China) was utilized and two clubbed platinum electrodes (diameter 1 mm, length 50 mm) were used in CE separations.

The main noise source in RBI detection appeared to be associated with temperature fluctuations. Since the RI of most liquids changes several parts per ten thousand with one degree temperature fluctuation, the detection limit of RBI, 10^{-6} ΔRI, will be corresponding to a temperature fluctuation of a few millidegrees. Therefore, before each sample load, a high voltage same as the separation voltage had been added to drive the electro-osmotic flow (EOF) without samples for at least 15 min, by which the joule heat would reach a balance. In addition, a fast sample injection was taken to alleviate the undulation from the absence of the high voltage.

2.3. Chemicals

All chemicals are analytical reagent grade or better. Amaranth, rhodamine 6G, dopamine, alanine and serine were purchased from Sigma–Aldrich Company (St Louis, MO, USA), while borax and ethanol were obtained from Sinopharm Chemical Reagent (Shanghai, China). Ultrapure water was employed to prepare buffers and sample solutions, and a number of dilutions for sample solutions were taken to evaluate their linear ranges. All buffers and samples were filtered before the use.

3. Results and discussion

3.1. Theoretical comparison for the RBI signal with and without thermal lens

Since owning different RI, different samples are corresponding to different RBI signals. However, coaxial TLS detection is based on the thermal lens phenomenon, which would induce the RI fluctuation at the detection spot. In this case, RBI signals with and without thermal lens were theoretically compared to ensure its reliability.

Franko and Tran reviewed analytical thermal lens instrumentations, and the temperature fluctuation in the thermal lens region was given as [37]:

$$\Delta T(x,y,t) = \frac{\ln(10)\alpha P}{\pi\rho C_p}\int_0^t \frac{1+\cos\omega t}{[8D(t-\tau)+\omega_e^2]} \times \exp\left[\frac{-2([x-v_x(t-\tau)]^2+y^2)}{8D(t-\tau)+\omega_e^2}\right] d\tau \qquad (1)$$

where the $T(x, y, t)$ is the time-dependent temperature distribution inside the sample, α is the decadal absorption of the medium per unit length, P is the power of the laser beam, ρ is the density, C_p is the specific heat at a constant pressure of the medium with a uniform velocity V_x in the x direction, ω is the angle frequency of the chopper, t is the time of excitation, and D equals to $k/\rho C_p$, in which k is the thermal conductivity.

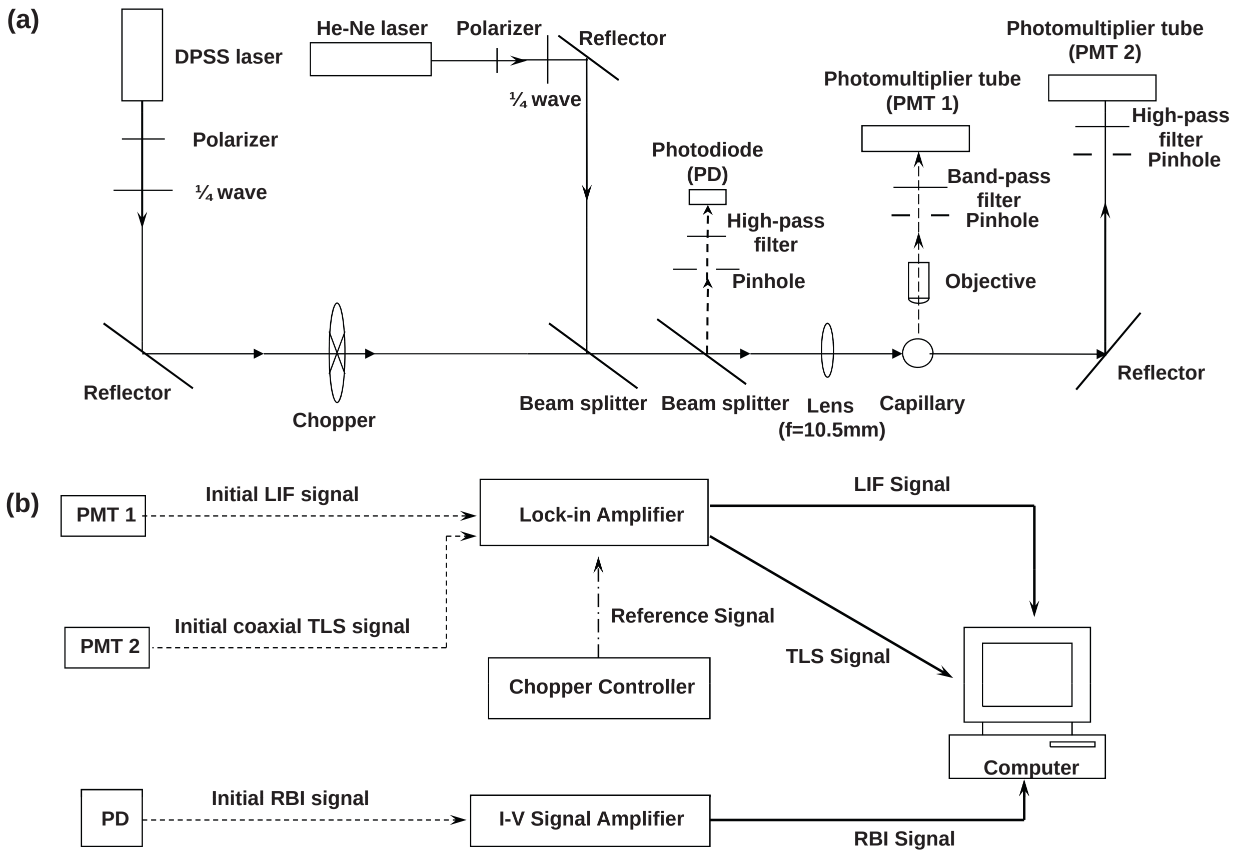

Fig. 1. The realization of simultaneous LIF, coaxial TLS and RBI detection for CE. (a) Optical schematic representation and (b) signal modulation block diagram.

Then, the RI change corresponding to the temperature fluctuation could be presented as:

$$\Delta n(x, y, t) = \left(\frac{\partial n}{\partial T}\right)_{T_A} \Delta T(x, y, t) \tag{2}$$

where $\left(\frac{\partial n}{\partial T}\right)_{T_A}$ is a constant, which stands for the RI change per unit temperature fluctuations.

In order to predigest the calculation, static samples were appointed to be evaluated. In fact, with the same thermal lens, static samples would result greater temperature fluctuations compared with flowing samples. Therefore, with the static sample, greater RI changes corresponding to the same temperature fluctuation would be obtained, based on which the invalidity of following evaluations could be ensured.

In accordance with our experimental setups and all pre-hypothesis, certain physical parameters could be obtained as following: $r_s = x^2 + y^2$, $P = 20\,\text{mW}$, $\alpha = 2.9 \times 10^{-4}\,\text{mm}^{-1}$, $k = 0.598\,\text{mW}\,\text{mm}^{-1}\,\text{K}^{-1}$, $\left(\frac{\partial n}{\partial T}\right)_{T_A} = 0.91\,\text{K}^{-1}$, $V_x = 0$, $\rho = 1.0 \times 10^{-3}\,\text{g}\,\text{mm}^{-3}$, $C_p = 4.184 \times 10^3\,\text{mJ}\,\text{K}^{-1}\,\text{g}^{-1}$ and $\omega = 502.65\,\text{S}^{-1}$. According to Eqs. (1) and (2) and preceding parameters, the RI change within two heating periods, at the detection spot of coaxial TLS, had been indicated in Fig. 2. At each time $t_{(s)}$, the exact centre of the thermal lens, corresponding to $r_s = 0$, showed the largest RI change. Therefore, when the probe laser beam in RBI detection is strictly collinear with the pump laser beam in coaxial TLS detection, the influence to the RBI detection will be the greatest. Therefore, the probe beam in RBI detection was assumed to be exactly collinear with the pump beam in TLS, which will ensure the reliability of following theoretical calculations.

In Fig. 2, RI changes within two heating periods were no more than 4×10^{-6}, which was rather faint. However, only when the ratio, between the RBI signal and its fluctuation from the thermal lens, was less than 5%, the influence of the thermal lens would be regarded negligible. Therefore, the preceding ratio was theoretical calculated.

By Li and Deng, the ideal theoretical optical matrixes of the RBI detection were given as [34,38]:

$$\begin{pmatrix} A_1 & B_1 \\ C_1 & D_1 \end{pmatrix} = \begin{pmatrix} 1 & L_2 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ \frac{-1}{f} & 1 \end{pmatrix} \begin{pmatrix} 1 & 2L_0 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ \frac{-1}{f} & 1 \end{pmatrix} \times \begin{pmatrix} 1 & L_1 \\ 0 & 1 \end{pmatrix} \tag{3}$$

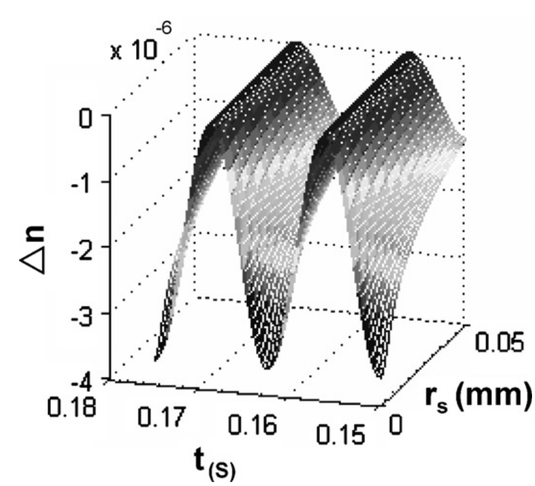

Fig. 2. The RI change within two heating periods.

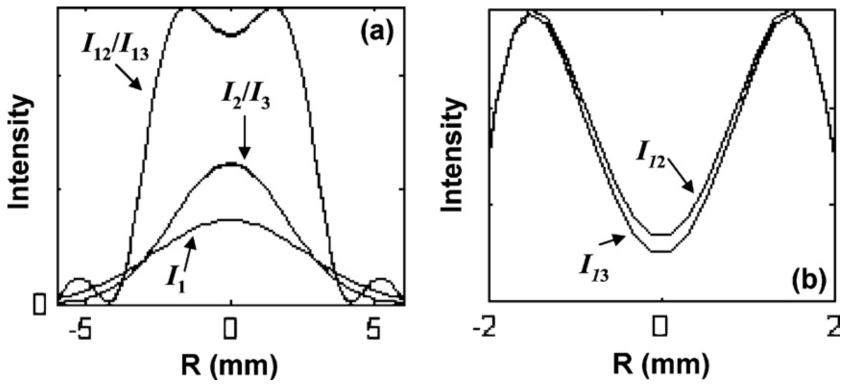

Fig. 3. The influence of the thermal lens to the RBI signal. (a) The full-scale graph and (b) the magnification for a part of full-scale graph.

$$\begin{pmatrix} A_2 & B_2 \\ C_2 & D_2 \end{pmatrix} = \begin{pmatrix} 1 & L_2 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ \frac{-1}{f} & 1 \end{pmatrix} \begin{pmatrix} 1 & L_0 \\ 0 & 1 \end{pmatrix} \times \begin{pmatrix} 1 & 2\left[\left(\frac{d-d_0}{n_0}\right)+\left(\frac{d_0}{n}\right)\right] \\ 0 & 1 \end{pmatrix} \times \begin{pmatrix} 1 & L_0 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ \frac{-1}{f} & 1 \end{pmatrix} \begin{pmatrix} 1 & L_1 \\ 0 & 1 \end{pmatrix} \quad (4)$$

where f is the focus length of the lens; L_0 denotes the distance from the focusing lens to the front outer surface of the capillary, which is about 10 mm in the proposed optical scheme; L_1 is the propagation distance, which is around 600 mm, from the laser beam waist to the focusing lens; L_2 denotes the distance from the lens to the detection plane, which is around 620 mm; d_0 and d are the inside and outside diameter of the capillary tube; n_0 is the RI of the tube wall.

Moreover, preceding two matrixes could be induced as:

$$\omega_i = \omega_0({A_i}^2 + {B_i}^2{Z_c}^2)^{1/2} \quad (i = 1, 2) \quad (5)$$

$$R_i = \frac{{A_i}^2{Z_c}^2 + {B_i}^2}{A_iC_i{Z_c}^2 + B_iD_i} \quad (i = 1, 2) \quad (6)$$

where ω_1 and ω_2 are the $(1/e^2)$ radii of two reflected beams at the detection plane, respectively; R_1 and R_2 are the radii of curvature of two reflected beams at the detection plane, respectively; $Z_c = \pi{\omega_0}^2/\lambda$ is the focal distance of the beam; ω_0 is the waist radius of the laser beam whose position depends on the configuration of the laser cavity.

Then, the light intensity on RBI detection plane could be given as:

$$I(r) = I_1(r) + I_2(r) + 2\sqrt{I_1(r)I_2(r) \times \cos}\left[k\left(\Delta + \frac{r^2}{2R_2} - \frac{r^2}{2R_1}\right)\right] \quad (7)$$

and

$$I(r) = \frac{TC}{\omega_1^2}\,\exp\frac{-2r^2}{\omega_1^2} \quad (8)$$

$$I_2(r) = \frac{TC(1-T^2)}{\omega_2^2}\,\exp\frac{-2r^2}{\omega_2^2} \quad (9)$$

$$\Delta = 2[nd_0 + n_0(d - d_0)] \quad (10)$$

where C is a constant; $k = 2\pi/\lambda$ is the wave number, λ is the laser wavelength; T is the reflectivity of the outer surface of the capillary; $r = (x^2 + y^2)^{1/2}$; $I_1(r)$ and $I_2(r)$ are the intensity profile of two beams reflected from front and rear capillary surfaces, respectively; Δ is the optical path difference between preceding two reflected beams, and n is the RI of the liquid in the tube.

According to Eqs. (3)–(10), RBI signals with and without thermal lens had been calculated for the same system as in Fig. 2. Corresponding results had been indicated in Fig. 3, in which I_1 was the intensity profile of the beam reflected from the front capillary wall, while I_2 and I_3 were intensity profiles of the beam reflected from the rear capillary wall without and with a thermal lens, respectively. Then, I_{12} and I_{13} were the RBI signal without and with a thermal lens, correspondingly. In Fig. 3(a), the difference between I_{12} and I_{13} was too faint to be observed, which indicated that the influence from the thermal lens was rather faint. Furthermore, when a part of Fig. 3(a) was magnified in Fig. 3(b), the difference between I_{12} and I_{13} was much less than 5% of the I_{12}, which was proved to be negligible.

3.2. Theoretical optimization for the lens focus

The focus length of the lens is another important parameter for the simultaneous LIF, coaxial TLS and RBI detection. In conventional LIF and coaxial TLS, a lens with short focus length is usually employed to focus the excitation laser beam and introduce it into the capillary. Generally, smaller laser waist will be obtained by a lens with a smaller focus length. However, RBI detection is strictly related to the focus length of the lens according to Eqs. (3)–(6), while there are no strict requirements for the lens used in LIF and coaxial TLS.

Deng and Li reported that the contrast of RBI fringe (ω_1/ω_2) was related with the focus length of the lens f [34]. In their report, when f was around 10 mm, the contrast of RBI fringe was closed to the highest value, and ω_1 and ω_2 were limited within a relatively small value, all of which ensured a satisfactory LOD. Furthermore, in accordance with our experimental results, an inappropriate L_0 will lead too many or too few fringes in the detection area, which may influence the sensitivity of the RBI detection. According to Eq. (7), the RBI fringe number in the detection area is related with the phase difference, which is closely interrelated with the optical path difference and the wave front difference for two reflected beams. Moreover, their optical path difference is a constant based on Eq. (10). Therefore, the fringe number in the detection plane is directly related with the focus length L_0, which determines the radii of curvature, and indirectly influences the wave front difference.

Based on preceding analysis, the fringe number in the detection area, whose radius was no more than 5 mm, was shown under different focus lengths in Fig. 4. It could be observed that a lens with a focus around 10 mm would result too many fringes, which would make its optical detection difficulty. Furthermore, if its focus length was around 9.8 mm or 10.2 mm, a sharp decrease of the fringe number would be realized, and fewer fringes should be better

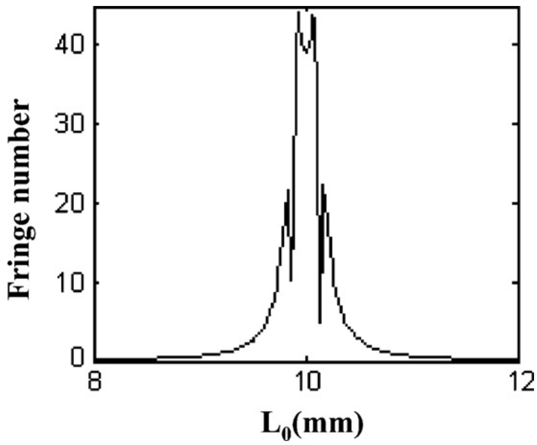

Fig. 4. The fringe number under different focuses for the key lens.

considering the optical realization for the RBI detection. In practical experiments, the fringe number was fewer with a 10.5 mm lens compared with its counterpart with a shorter or longer focus, all of which was similar with Fig. 4. It was worthy to emphasize that a 10.5 mm focus length would also be helpful to realize an acceptable RBI fringe contrast [34]. Based upon all preceding results, a 10.5 mm lens was chosen to be the key lens in the proposed detection.

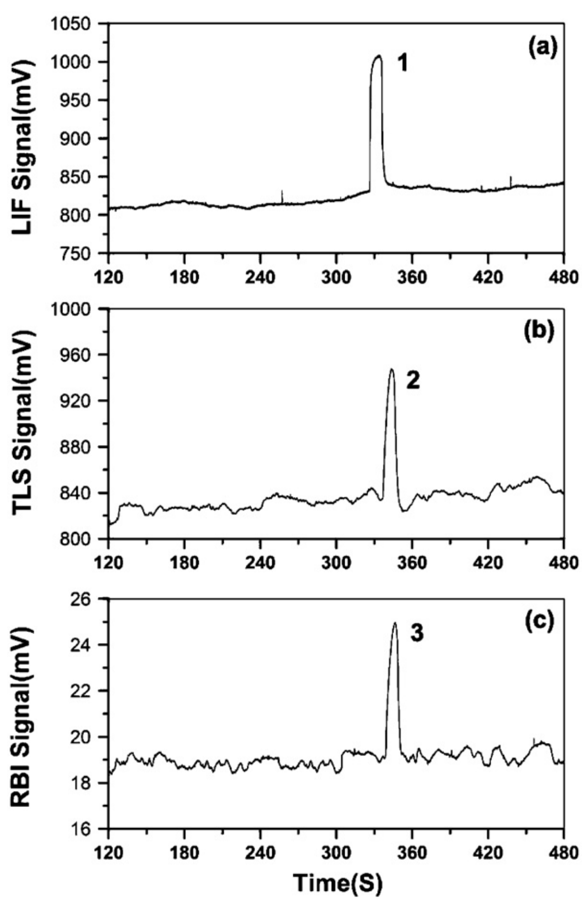

Fig. 5. The CE identification of rhodamine 6G by simultaneous LIF, coaxial TLS and RBI detection. Capillary: 40 cm (33 cm effective length) × 75 μm I.D. Buffer: 30 mM borate buffer, pH 9.5. Separation voltage: 12 kV voltage. Injection: 0.1 psi for 4 s. Peaks: (1) rhodamine 6G (8.4×10^{-8} M), (2 and 3) electroosmotic flow (EOF, marked by 0.1% ethanol).

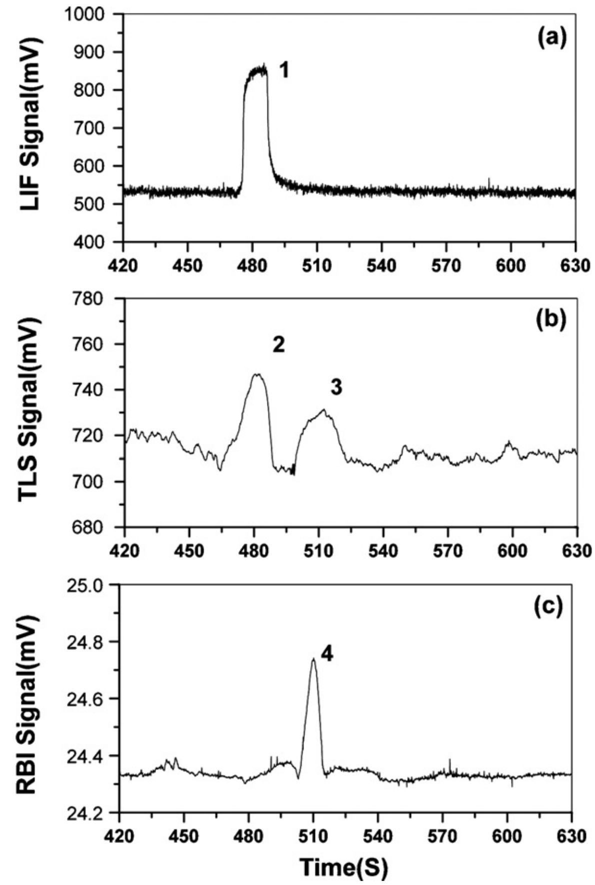

Fig. 6. The CE identification of rhodamine 6G by simultaneous LIF, coaxial TLS and RBI detection. Capillary: 45 cm (38 cm effective length) × 75 μm I.D. Buffer: 30 mM borate buffer, pH 9.5. Separation voltage: 12 kV voltage. Injection: 0.1 psi for 4 s. Peaks: (1 and 2) rhodamine 6G (4.2×10^{-5} M), (3 and 4) electro-osmotic flow (EOF, marked by 0.1% ethanol).

3.3. Simultaneous LIF, coaxial TLS and RBI detection for CE

In the first place, 8.4×10^{-8} M rhodamine 6G was employed to evaluate the LIF detection in the proposed detection, which was illustrated in Fig. 5. It should be pointed out that Fig. 5(a)–(c) was all collected simultaneously. In Fig. 5(a), a baseline separated peak for rhodamine 6G was obtained, which suggested a satisfactory sensitivity. Meanwhile, Fig. 5(b) and (c), corresponding to coaxial TLS detection and RBI detection, indicated an EOF peak with the same retention time, both of which stood for a satisfactory work status. Based on Fig. 5, it was abundant to draw the conclusion that the LIF detection was still of high sensitivity in the proposed detection.

Then, 4.2×10^{-5} M rhodamine 6G, which could be observed in both LIF detection and coaxial TLS detection, was analyzed by the reported detection scheme. In Fig. 6(a) and (b), a baseline separated peak for rhodamine 6G could be observed at the same retention time, and this result approved the reliability of LIF and coaxial TLS detection in the reported detection. Moreover, the amaranth which was more sensitive in TLS detection was appointed to evaluate its performance. It was worth mentioned that the LOD of TLS detection was worse than LIF detection in the reported detection. Therefore, in order to observe the peak for rhodamine 6G in both coaxial TLS detection and LIF detection in Fig. 6, the sensitivity of LIF detector (PMT 1) had been depressed by controlling its power supply to 60% full output. In accordance with Fig. 6, the reliability of the coaxial TLS detection in the proposed detection could be ensured.

Fig. 5 indicated the high sensitivity of LIF detection, and the LIF detector had been adjusted to the highest sensitivity. Therefore, a

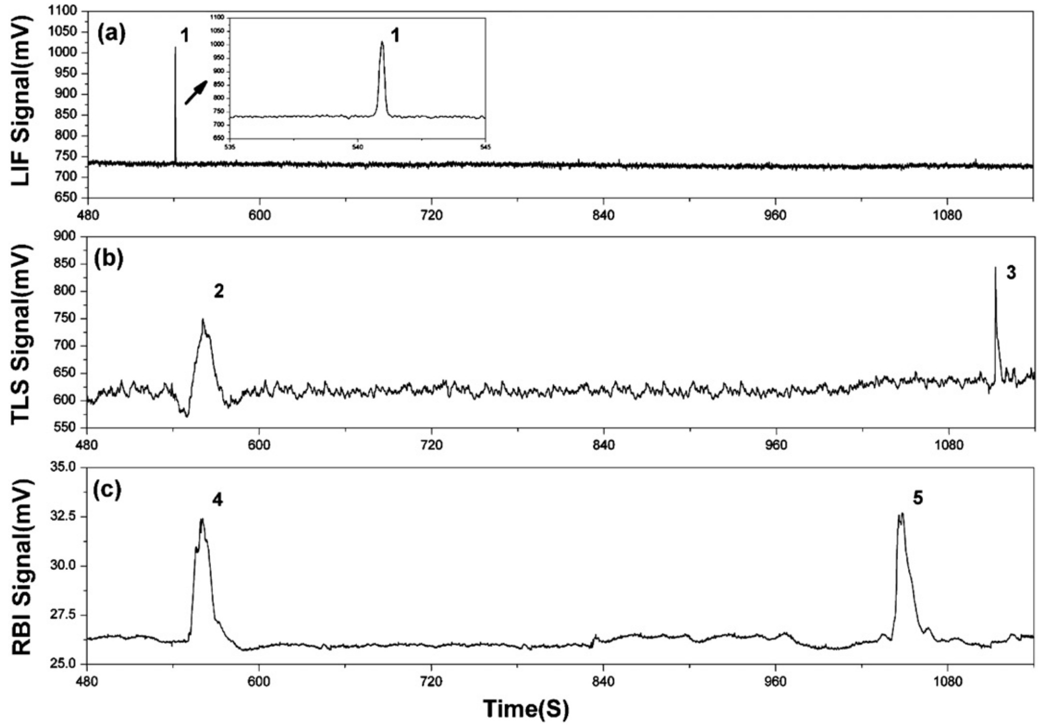

Fig. 7. The CE identification of rhodamine 6G, amaranth and serine by simultaneous LIF, coaxial TLS and RBI detection. Capillary: 48 cm (41 cm effective length) × 75 μm I.D. Buffer: 30 mM borate buffer, pH 9.5. Separation voltage: 10 kV voltage. Injection: 0.1 psi for 4 s. Peaks: (1) rhodamine 6G (1.4 × 10^{-8} M), (2 and 4) electro-osmotic flow (EOF, marked by 0.1% ethanol), (3) amaranth (2.6 × 10^{-7} M), and (4) serine (8.1 × 10^{-5} M).

volume overload phenomenon appeared in Fig. 5(a), while the concentration for rhodamine 6G was comparatively low. In Fig. 6, it was aimed to indicate the availability of both coaxial TLS detection and LIF detection. In this case, the rhodamine 6G was still over loaded in Fig. 6(a), though its sensitivity had been depressed. When the LIF sensitivity is adjusted correspondingly and the sample concentration is suitable, the over load phenomenon could disappear.

Under conventional LIF detection for CE, amino acids need to be derived [39], which will frequently make the reagent for the derivatization much overloaded compared with amino acids. However, if derivatization reagents such as FITC or rhodamine isothiocycanate are with self-fluorescence, the peak for those reagents will be much greater than derived amino acids, which will frequently be overlapped or even completely covered up. Unfortunately, those derivatization reagents are all widely utilized. On the contrary, dopamine, alanine and serine were determined without any derivatizations in Supporting Fig. 1. The result supported the versatile property of the reported detection scheme.

Furthermore, the simultaneous CE identification of rhodamine 6G, amaranth and serine with the simultaneous LIF, coaxial TLS and RBI detection was demonstrated in Fig. 7. Rhodamine 6G, amaranth and serine without any derivatizations had been determined in corresponding detections, all of which indicated the combination of high sensitivity and universal property.

3.4. LOD, linear ranges and RSD

Based upon preceding determinations, the proposed detection had been evaluated. The LOD for the rhodamine 6G in LIF detection was 5.1 × 10^{-9} M, and its linear range reached over 3 orders. In addition, the LOD for amaranth in coaxial TLS detection was 3.2 × 10^{-8} M, and its linear range reached 2.5 orders. Moreover, the LOD for serine in RBI detection was 6.6 × 10^{-6} M, and its linear range achieved near 3 orders. The last, the RSD in LIF, coaxial TLS and RBI was below 10%, which was an acceptable value for CE determinations. All RSDs were based upon the peak area in those analyses, whose experimental parameters were the same as Figs. 5–7. Its performance in RBI detection was better than reported RBI detection system [34], while its performance in LIF and coaxial TLS detection was no worse than former research reports [33].

4. Conclusions

We have developed a novel multifunctional CE detector which accomplished simultaneous LIF, coaxial TLS and RBI detection. By the utilization of a DPSS laser and a He–Ne laser, the reported detection had been realized. In addition, RBI signals with and without a thermal lens had been theoretically compared, based on which its reliability had been ensured. Furthermore, the focus length of the lens had been optimized to improve its performance. The last but not least, several samples had been determined to indicate its own advantages, as well as to indicate no worse performances compared with former reports.

Acknowledgements

This research is financially supported by National Natural Science Foundation of China (No. 20427002, No. 90409013, No. 30772058). Great thanks to Mr. George Hamilton and Mrs. Anna Stone (South-Central University for Nationalities, China) for their help in the English usage.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.talanta.2011.10.027.

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