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Research Article

A phenylboronic-acid functionalized liquid photonic crystal reagent for convenient and reliable detection of glucose

Yanxuan Zhao[1,§], Huimin Zhu[1,§], and Jianping Ge[1,2]✉

[1] *State Key Laboratory of Petroleum Molecular & Process Engineering, Shanghai Key Laboratory of Green Chemistry and Chemical Processes, School of Chemistry and Molecular Engineering, East China Normal University, Shanghai 200062, China*
[2] *Institute of Eco-Chongming, Shanghai 202162, China*
[§] *Yanxuan Zhao and Huimin Zhu contributed equally to this work.*

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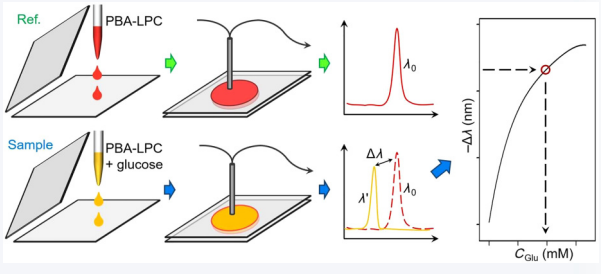
ABSTRACT: Photonic crystal sensing is an emerging technique that directly indicates the physicochemical changes with the change of structural color. However, there are still challenges in material synthesis, ease of use, and reproducibility of detection for traditional photonic crystal (PC) sensors. Here, a phenylboronic-acid functionalized SiO_2 liquid photonic crystal (PBA-LPC) reagent was developed for reliable detection of glucose concentration. It is convenient to prepare/use the LPC reagent because people only need to mix the responsive substance or the analyte solution with the LPCs in synthesis/detection. The sensing was based on the mechanism that the mixing of PBA-LPC reagent with glucose could release H^+ cations from PBA, which inhibited the deprotonation of the silanol group, weakened the particle surface charge, and blue-shifted the reflection of LPC. The PBA-LPC reagents responded to the glucose in different concentration ranges depending on the dosage of PBA, which ensured both a broad range detection and an accurate detection within a specific range. Meanwhile, it reported reproducible results due to the precise introduction of PBA and its sufficient interaction with glucose. Furthermore, the PBA-LPC reagent showed a selective response to glucose and good anti-interference capability against the presence of NaCl, $CaSO_4$, KH_2PO_4, and Vitamin C. Due to these properties, the PBA-LPC reagent could serve as a new material for blood sugar tests and it also demonstrated the great potential of LPCs in sensing applications.



KEYWORDS: phenylboronic-acid, liquid photonic crystal, detection reagent, glucose sensing

1 Introduction

Responsive photonic crystals have attracted great interest due to their potential application in colorimetric sensors [1], display units [2, 3], photonic printing [4–6], anti-counterfeiting materials [7–9], optical devices [10, 11], and smart skins [12, 13]. Among these applications, photonic crystal (PC) sensors were broadly investigated because the responsive PC tightly connected the external stimuli with the photonic structure so that the change of structural color could indicate the change of physical [14–17] or chemical [18–21] parameters. As a typical example, a PC-based glucose sensor was proposed for the diagnosis and therapy of diabetes disease. It provided a fast and convenient way to indicate the blood sugar levels through the color change of the PCs.

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✉Address correspondence to jpge@chem.ecnu.edu.cn

Like most reported PC sensors, the glucose sensors [22–26] were usually fabricated by introducing a glucose-responsive substance to the PC structures, such as PC hydrogels and inverse opals. For instance, Asher et al. [27] have developed a 5-amino-2-fluorophenylboronic acid bonded acrylamide PC hydrogel and demonstrated its application in the sensing of glucose with concentrations in the blood. As the glucose concentration increased, the PC hydrogel was more tightly cross-linked through the formation of a boronate-glucose complex, which shrank the PC hydrogel and blue-shifted the reflection signal. In another instance, Meng et al. [28] prepared a molecularly imprinted PC sensor by filling the voids of a poly(methyl methacrylate) colloidal crystal with a poly(acrylamide-acrylate) hydrogel covalently bonded with the boronate-glucose complex, followed by the removal of colloidal particles and glucose molecules. The molecularly imprinted hydrogel network swelled upon the adsorption of glucose from the analyte solution, which led to the volume expansion of the inverse opal gel and redshift of the reflection peaks.

Although solid [29–32] or hydrogel [22–26, 33–35] PC sensors

have been demonstrated for glucose sensing in different material systems, there are still challenges in sensor preparation and analyte detection for these traditional PC sensors. First of all, the fabrication of a PC sensor usually requires multiple steps, including colloidal assembly, hydrogel polymerization, and modification by responsive substances. Each step needs to be carefully controlled to achieve a completely equivalent sensing material. Secondly, the reproducible response could be a concern in parallel detections as the reflection signal was affected by the loading amount of the responsive groups, the fixation of the polymeric PC on the substrate, and even the testing position in the material.

Compared to the traditional PC sensor, liquid photonic crystal (LPC) was an emerging detection reagent with superior performance for sensing in a liquid environment. Because the LPC was sensitive to many physicochemical parameters, such as pH value, ionic strength, solvent polarity, temperature, viscosity, etc. It was easy to prepare an LPC detection reagent as people only needed to mix the responsive substance with a premade LPC. It was also convenient to use the LPC reagent, where the reflection measured before and after mixing the analyte with the LPC could be compared to achieve the sensing results. More importantly, the LPC reagent could quickly and quantitatively interact with the analyte in solution, which ensured an accurate and repeatable sensing result.

In this work, a phenylboronic acid (PBA) functionalized SiO_2 liquid PC reagent was developed for reliable detection of glucose concentration. PBA and its derivatives had been widely studied for the sugar sensors because of their selective binding with 1,2- and 1,3-diols to form boronate esters. Such non-enzymatic sensors addressed the problem of instability for the conventional biosensors based on glucose oxidase. The selective binding forms negatively charged PBA-sugar adducts in neutral environment, which effectively changed the ultraviolet (UV)–visible and fluorescence spectra of the PBA derivatives. When the electrodes or PC hydrogel were modified with PBA derivatives, the selective binding also changed the electrochemical behavior of the electrode or the optical property of the hydrogel, which turned them into potentiometric or structural-colored glucose sensors [36]. Illuminated by these successful applications, we developed a new PBA-LPC sensing agent. The sensing was based on the following mechanism that the mixing of PBA-LPC reagent with the glucose solution could release H^+ cations from PBA, which inhibited the deprotonation of the silanol group, weakened the particle surface charge, and blue-shifted the reflection signal of LPC. Then, parallel experiments were performed to investigate the influence of PBA dosage on the concentration range, the response sensitivity (RS), and the accuracy of glucose detection. The advantages of PBA-LPC reagent in reproducibility and glucose selectivity were also demonstrated by experimental results. Since the detection range (DR) fully covered the glucose concentration in human blood, we also studied the anti-interference capability of the PBA-LPC reagent against the ions and Vitamin C in blood and screened the most suitable reagent from the PBA-LPCs composed of different solvents.

2 Experimental section

2.1 Materials

Aqueous ammonia ($NH_3{\cdot}H_2O$, 28%), tetraethyl orthosilicate (TEOS, 98%), ethanol (EtOH, 99.9%), 1,4-butanediol (BDO, 99%), dimethyl sulfoxide (DMSO, 99%), and glucose were purchased from China National Pharmaceutical Group Co. Ltd. (Sinopharm). Ethylene glycol (EG, 99%) and triethylene glycol (TEG, 99%) were purchased from Belling Technology Co. Ltd. Propylene carbonate (PCb, 99%), PBA (97%), fructose, xylose, D-galactose, D-sucrose, and chitosan were purchased from Aladdin Bio-Chem Technology Co. Ltd. All chemicals were used directly as received without further purification.

2.2 Synthesis of PBA functionalized SiO_2/EG liquid PC reagent

SiO_2 colloidal particles were first synthesized by a modified Stöber method at room temperature. TEOS (16 mL) was injected into the mixture of ethanol (200 mL), H_2O (14 mL), and $NH_3{\cdot}H_2O$ (8 mL), which produced SiO_2 particles with a diameter of 180 nm after a 4-hour reaction. The SiO_2 particles were collected by centrifugation, washed with ethanol 3 times, and dried in vacuum for storage. For the preparation of PBA (0.01 mM) functionalized SiO_2/EG liquid PCs, SiO_2 particles (71.4 mg) were first dispersed in ethanol (1.1 mL), and then mixed with EG (65 μL) and an ethanol solution of PBA (0.1 mM, 10 μL) to form a homogeneous colloidal solution under sonication. After being placed in an oven at 90 °C for 2 h, all the ethanol evaporated and the solution turned to a supersaturated colloidal solution with a total volume of 0.1 mL and particle volume fraction of 35%. When the solution was kept standing for several minutes, the particles spontaneously precipitated to form SiO_2/(PBA-EG) LPC. For the synthesis of SiO_2/(PBA-EG) LPCs with PBA concentrations of 0.005, 0.1, and 1.0 mM, the synthetic procedures were the same except that a different amount of PBA was added at the beginning. For the synthesis of other SiO_2/(PBA-solvent) LPCs, the synthetic procedures were also the same except that EG was replaced by PCb, BDO, DMSO, and TEG.

2.3 Working curves for the detection of glucose

Firstly, a small amount of the SiO_2/(PBA-Solvent) LPC reagent was sandwiched by two glass sides with interspacing of 180 μm to form a thin liquid PC film in 5 min, whose reflection wavelength was measured to be λ_0. Then, the LPC reagent was mixed with "standard glucose solution" with different concentrations (C_{Glu}), whose reflection wavelengths were measured to be λ_1, λ_2, …, etc. Finally, the working curves for the glucose detection could be plotted by the C_{Glu} and the corresponding wavelength change ($\Delta\lambda_1$, $\Delta\lambda_2$, …) compared to the LPC reagent.

2.4 Glucose detection by SiO_2/(PBA-EG) reagent

For a typical detection of glucose concentration, 10 μL of the glucose solution was first mixed with 100 μL of the SiO_2/(PBA-EG) LPC reagent. A small amount of the mixing solution was sandwiched by two glass sides to form a thin liquid PC film in 5 min, whose reflection wavelength was measured to be λ'. According to its difference to the wavelength of the LPC reagent, one could determine the glucose concentration from the "$\Delta\lambda$–C_{Glu}" working curve.

2.5 Characterizations

The reflection spectra were recorded by an Ocean Optics Maya 2000 Pro spectrometer coupled to a six-around-one reflection probe, where both the incident and reflective angle were fixed at 0°. The morphology and particle size of SiO_2 colloidal particles were characterized by the Hitachi S4800 scanning electron microscopy (SEM). The optical microscopy images were collected by an

Olympus BXFM reflection-type microscopy operated in dark-field mode. The zeta potential of SiO_2 particles in the solution was measured by Malvern Zetasizer Nano ZS90 at room temperature.

3 Results and discussion

LPC composed of SiO_2 particles and various solvents with high boiling points were prepared by an evaporation-induced supersaturation and particle assembly process [37]. In the synthesis, SiO_2 particles could be well dispersed in an ethanol-based mixing solvent. The selective evaporation of ethanol drove the precipitation of SiO_2 particles from the left solvent, such as EG, which facilitated the spontaneous colloidal assembly to form the LPCs. This method was a universal way to prepare SiO_2-based LPC with high-boiling-point solvents, such as SiO_2/EG, SiO_2/PCb, SiO_2/BDO, SiO_2/DMSO, and SiO_2/TEG LPCs (Figs. 1(a) and 1(b)).

The as-prepared colloidal solution presented typical characteristics of LPCs. For example, the LPC was usually composed of two phases, which were solvent-wrapped colloidal crystals and amorphous colloidal dispersion in solution. Such crystalline and amorphous assembly structures could be directly observed in the microscopy image as colorful and colorless domains, and they could also be confirmed by the ordered and disordered particle arrangement in the SEM images (Figs. 1(c)–1(e)). The LPC was a fluid with photonic bandgap and structural colors. It was also a metastable structure, which renders it reversible assembly and disassembly characteristics. Generally, the LPC could be assembled in several minutes without external disturbance, and it disassembled into a supersaturated colloidal solution in several seconds under a weak disturbance. The periodic change of the reflection intensity suggested that the assembly and disassembly were fully reversible (Figs. 1(f)–1(h)).

The LPC solution was suitable for constructing a liquid-form detection reagent due to its sensitive response to broad physicochemical conditions in solution and its unique response mechanism that well integrated the sensing process and the signal output. On one hand, the assembly structure of the LPC was greatly affected by the physical properties and chemical environment of the colloidal solution, which meant any change in particle concentration, pH value, solvent polarity, and viscosity would change the LPC's bandgap and reflection. Responsive substances could be directly added to render the LPC responsive properties. Therefore, the relationships between stimuli, LPC structure, and optical signals made the LPC promising sensing materials. On the other hand, the reversible assembly and disassembly characteristic gave LPC a unique sensing mechanism. In the disassembly state, analytes could be well mixed with the LPC reagents so that a full interaction and an equilibrium state could be achieved quickly.

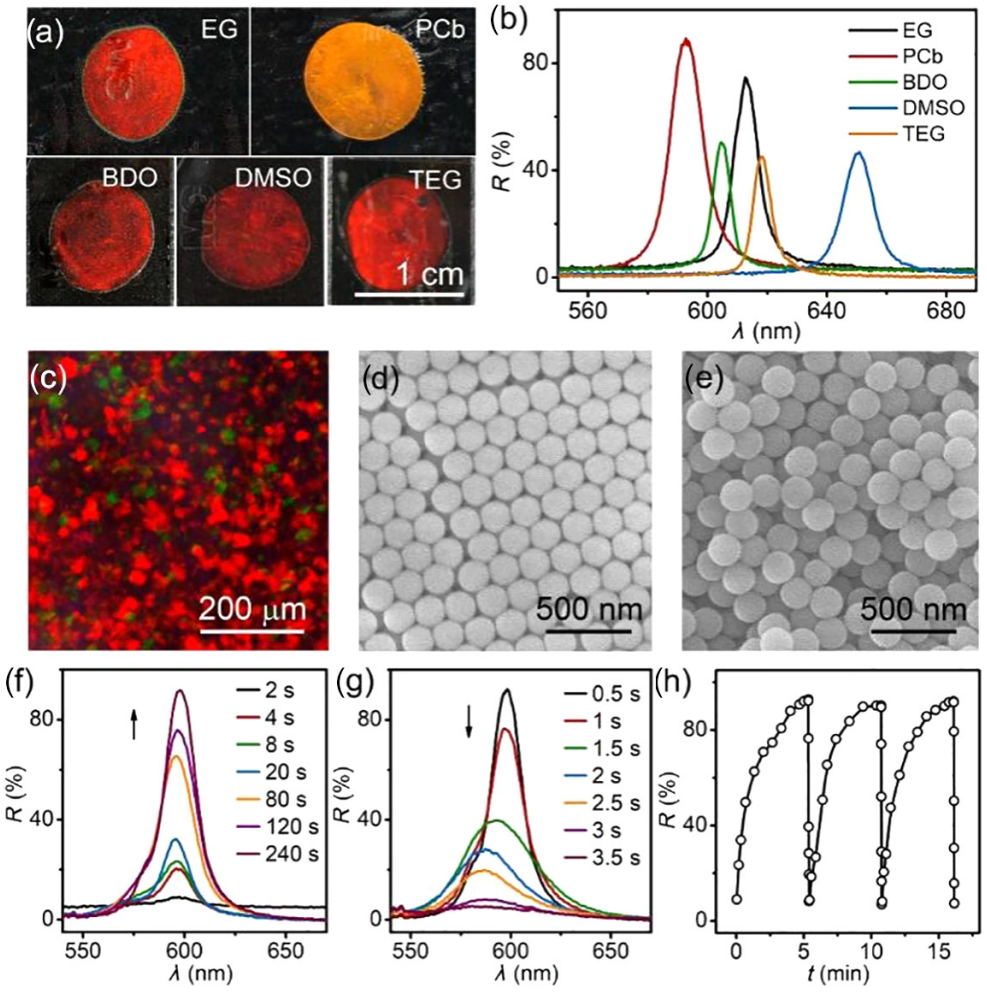

Figure 1 Characteristics of SiO_2/solvent LPCs. (a) and (b) Digital photos and reflection spectra of LPCs prepared by SiO_2 particles and different solvents; (c)–(e) optical microscopy and SEM images of SiO_2/EG LPCs composed of crystalline and amorphous colloidal assembly structures; (f)–(h) reversible assembly and disassembly proved by the periodic change of the reflection intensities.

While, in the assembly state, the LPC showed the reflection signals responding to the external stimuli, which was then used to identify the physical/chemical changes. In the following discussion, a PBA functionalized SiO_2/EG LPC for the detection of glucose concentration (C_{Glu}) in aqueous solution was developed to demonstrate this kind of new sensing material.

The PBA functionalized SiO_2/EG LPC, abbreviated as the SiO_2/(PBA-EG) reagent, showed a sensitive response to the glucose in solution, where the reflection peak blue shifted with the increase of glucose concentration (Figs. 2(a)–2(c)). In a typical sensing process, the SiO_2/(PBA-EG) LPC reagent was used as the reference sample, which showed a reflection peak at 594 nm. When the reagent was mixed with an aqueous solution of glucose with C_{Glu} from 0.1 to 10 mM, its reflection peak blue shifted from 4 to 48 nm, accordingly. Through the collection of adequate information, one could plot a working curve for the detection of C_{Glu} by the reflection wavelength change ($\Delta\lambda$). It was interesting to find that the reflection kept blue-shifting even the glucose exceeded the stoichiometric amount (red point) for reacting with the PBA in the ratio of 1:1. For the SiO_2/(PBA-EG) reagent with 0.01 mM of PBA, the trend of reflection blueshift remained until the glucose/PBA ratio reached 20:1. This was because the reflection change of the LPC reagent was related to the esterification of PBA with glucose. The esterification was an equilibrium reaction with a small equilibrium constant, which required a large amount of glucose to fully consume the PBA molecules. Such an equilibrium reaction was favorable to the sensing, as a few amounts of PBA component could detect glucose concentration with a broad range. It should be noted that the standard glucose solutions have pH values ranging from 6.0 (0 mM) to 3.8 (10 mM), which was different from that of the human blood (7.4). Fortunately, the reflection blueshift caused by them was close because only a small amount of glucose solution was added to the LPC reagent (Fig. S1 in the Electronic Supplementary Material (ESM)).

Further study on the sensing mechanism proved that the optical response was realized based on the intrinsic relationship between the release of H^+, the weakening of the particle surface charge, and then the lattice compression of the LPCs. When the glucose solution was mixed with the PBA-LPC reagent, a boronic ester formed along with the release of H^+ into the solution [38]. The released H^+ inhibited the deprotonation of the silanol group (Si-OH) on the surface of the SiO_2 particle and weakened its surface charge. Since the electrostatic repulsion between SiO_2 particles was a major factor influencing the LPC structure, the decrease of surface charge would eventually lead to the compression of the crystal lattice and then the blueshift of the reflection peak. To confirm the above mechanism, we investigated the influence of C_{Glu} upon the pH value for the diluted mixture of PBA-LPC and glucose as well as the interparticle spacing calculated from the reflection spectra (Figs. 2(d) and 2(e)). Along with the increase of C_{Glu}, we did observe the decrease of pH value and the shrinkage of the LPC lattice, which proved the delivery of responses and confirmed the glucose-induced reflection blueshift.

Based on the above sensing mechanism, we developed a

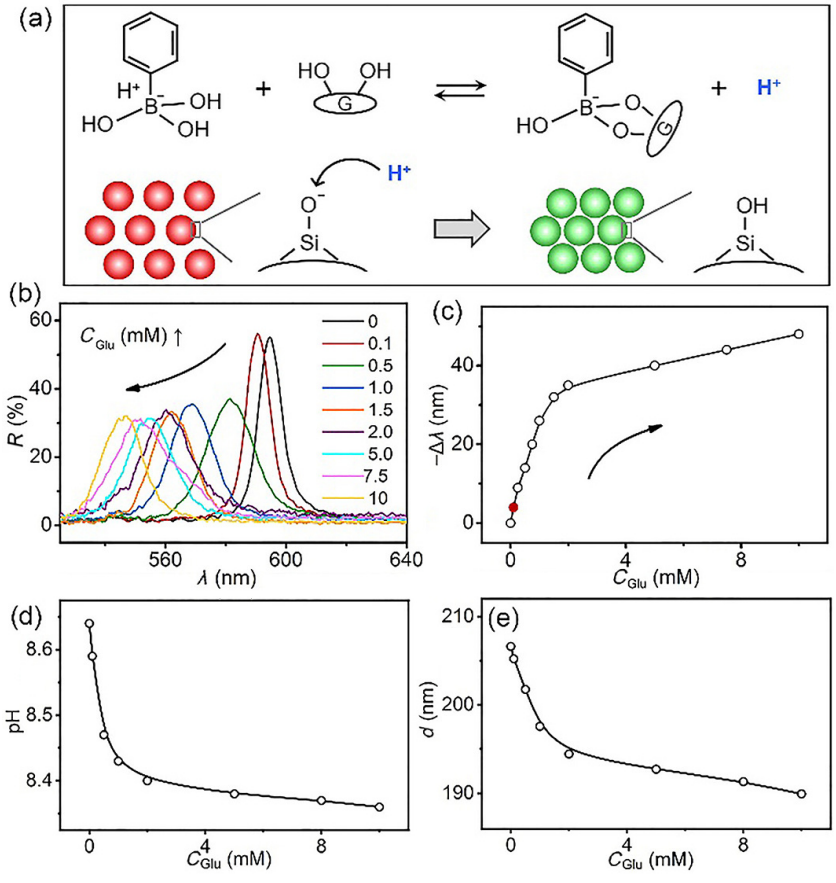

Figure 2 Working mechanism of LPC reagent for glucose detection. (a) Esterification of PBA with glucose led to the release of H^+, the decrease of surface charge, and the compression of crystal lattice; (b) and (c) the reflection of the SiO_2/(PBA-EG) LPC gradually blue-shifted when it was mixed with a glucose solution with increasing concentrations; evolution of (d) the pH values and (e) the interparticle spacing with the increasing glucose concentration.

convenient method to detect the glucose concentration in solution by using the SiO_2/(PBA-EG) LPC reagent (Fig. 3). As mentioned above, the working curve of "$\Delta\lambda$–C_{Glu}" was plotted after the reflection measurement of the mixture of the standard samples and the LPC reagent. For a typical detection, the reflection wavelength of the PBA-LPC reagent was first measured to be λ_0. Meanwhile, a small amount of glucose solution with an unknown concentration was mixed with the PBA-LPC reagent, whose reflection wavelength was measured to be λ'. Finally, the C_{Glu} could be determined by placing the reflection blueshift into the working curve.

Since the glucose detection was based on the esterification of boronic acid, the PBA dosage in the PBA-LPC reagent was found critical to the detection range, the response sensitivity, and the detection accuracy. Here, we prepared four PBA-LPC reagents with a PBA dosage of 0.005, 0.01, 0.1, and 1 mM and recorded their responses to the glucose with different concentrations (Fig. 4 and Fig. S2 in the ESM). The results were plotted in the "$\Delta\lambda$–C_{Glu}" curves, and all of them showed the same trend that the reflection peak blueshift along with the increase of glucose concentration. Through the curves, one could determine the detection range of C_{Glu}, the response sensitivity by the slope of the curve, and the detection error (DE) in a specific range. Taking the PBA(0.01 mM)-LPC reagent as a typical example, the detection range for C_{Glu} was determined to be 0–10 mM. The response sensitivity was found to be 21.4 nm/mM around the stoichiometry point of glucose/PBA (1:1) and it decreased to 1.78 nm/mM at higher C_{Glu}. The inflection point in the "$\Delta\lambda$–C_{Glu}" curves was caused by the dramatic change in response sensitivity between the high C_{Glu} and low C_{Glu} conditions. Due to the small equilibrium constant of the esterification of boronic acid, the change of H^+ concentration was remarkable at low

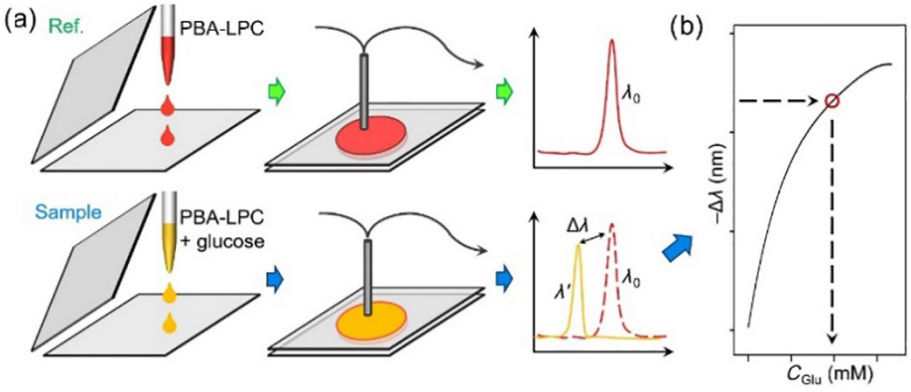

Figure 3 Detection procedures. (a) Measurement of reflection wavelength for the PBA-LPC reagent and its mixture with the glucose analyte; (b) determination of glucose concentration from the "$\Delta\lambda$–C_{Glu}" working curve.

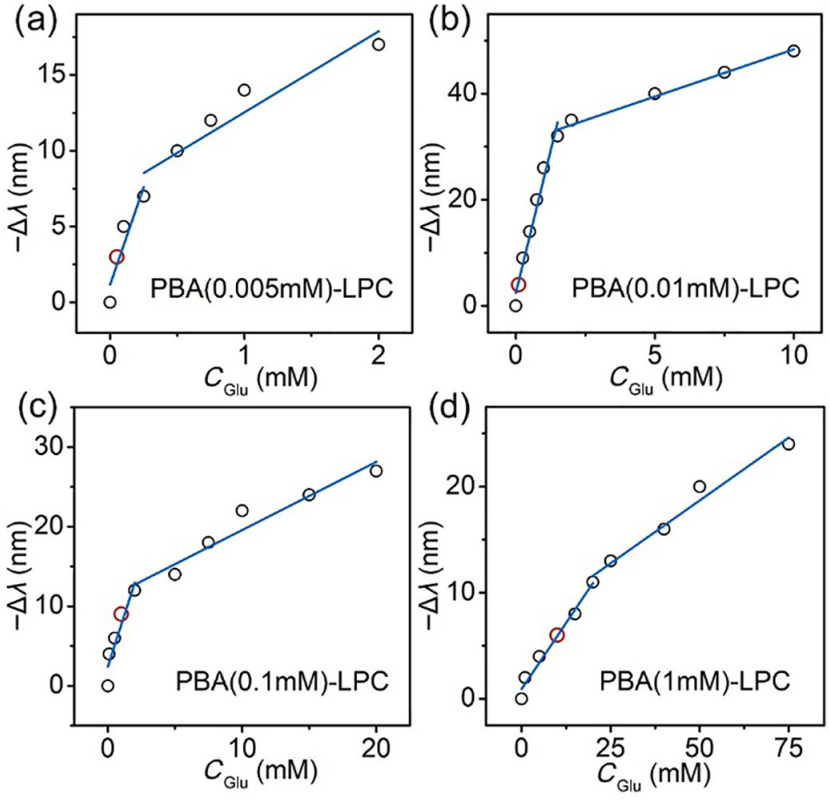

Figure 4 Detection range, response sensitivity, and detection accuracy. The evolution of the reflection blueshift of four SiO_2/(PBA-EG) LPCs when each of them was mixed with the glucose solutions with increasing concentrations. The PBA dosage in four LPCs was (a) 0.005 mM, (b) 0.01 mM, (c) 0.1 mM, and (d) 1 mM, respectively.

C_{Glu}, which led to the sensitive change of LPC's lattice and reflection. However, the release of H^+ at high C_{Glu} driven by the chemical equilibrium was gentle so the optical response became insensitive. If the error of spectra measurement was ±1 nm, the detection errors in two ranges were calculated to be ±0.0467 and ±0.562 mM, respectively. According to the parallel experiments of different PBA-LPC reagents, it could be concluded that the detection range amplified, the response sensitivity decreased, and the detection accuracy decreased when more PBA were added to the LPC (Table 1). It also suggested that such PBA-LPC reagent was compatible with glucose detection in a wide range with adequate accuracy. Considering the glucose concentration of human blood was 3–10 mM, the PBA(0.01 mM)-LPC reagent could be a potentially useful material for blood sugar tests.

Table 1 DR, RS, and DE in a condition of ±1 nm spectra error for the SiO_2/(PBA-EG) LPC reagent with different PBA dosages (C_{PBA})

C_{PBA} (mM)	DR (mM)	RS_1 (nm/mM)	RS_2 (nm/mM)	DE_1 (mM)	DE_2 (mM)
0.005	0–2	25.7	5.34	±0.0389	±0.187
0.01	0–10	21.4	1.78	±0.0467	±0.562
0.1	0–20	5.28	0.857	±0.189	±1.17
1	0–75	0.499	0.237	±2.00	±4.22

The PBA-LPC reagent showed repeatable responses and reliable detection results in the parallel tests due to the precise introduction of a responsive substance (PBA) and its sufficient interaction with the analyte (glucose). Here, the PBA(0.01 mM)-LPC reagent was used to test the glucose solution with fixed concentrations of 5 and 10 mM for 12 times. The reflection changes were almost the same among the parallel tests (Fig. 5(a)). For comparison, we also prepared traditional PC sensors, including the PBA-modified PC gel and the PBA imprinted PC structure, to detect the glucose concentration. Their sensing results showed relatively larger deviations (Fig. S3 in the ESM). We believed that good reproducibility first originated from the precise and quantitative loading of the responsive substance. PBA was simply mixed with the LPC reagent instead of being immobilized to the PC structure, which had less challenge in the preparation of sensing material. Furthermore, the LPC detection made sufficient contact and interaction between the responsive substance and the analyte through liquid mixing, which avoided inconsistent response as far as possible.

The PBA-LPC reagent also showed a selective response to glucose among various sugar molecules (Figs. 5(b) and 5(c)). Here, the aqueous solution of six sugar molecules including glucose, fructose, xylose, galactose, sucrose, and chitosan was mixed with the PBA-LPC reagent, whose reflection signals were compared with that of the PBA-LPC reagent. The reflection blue-shifted for glucose only. However, the reflection for the other sugars red-shifted due to the absence of PBA esterification and the redshift was caused by the dilution of LPC after mixing. The above results suggested a highly selective response of the PBA-LPC to the glucose because the furan structure of the glucose molecule had two pairs of cis-diols groups [39, 40] and each of them could form a stable cyclic boronic ester with PBA, leading to the release of H^+ and blueshift of the reflection signals.

Since the PBA-LPC reagent had the potential for blood sugar tests, we were interested in whether the SiO_2 particles assembled and the detection reagent functioned well under the interference of ions and nutrients in the blood. To realize the detection, plasma could be separated from all the blood cells. However, the plasma was still a complicated fluid composed of 90% of water, 7% of proteins, and 2% of other chemicals, including lipids, glucose, urea, and electrolytes. Among these chemicals, the electrolyte was obviously an interference to the colloidal assemblies, and the rest components might also bring influences. Limited by the research experience and lab conditions, we only investigated the influence of NaCl, $CaSO_4$, KH_2PO_4, and Vitamin C in this work, where most concentrations were set at the typical values in the blood. Previous studies about the liquid PCs suggested that LPCs constructed by different solvents had either strict or easing requirements for colloidal assembly, which reminded us to address the interference problem with a proper LPC reagent.

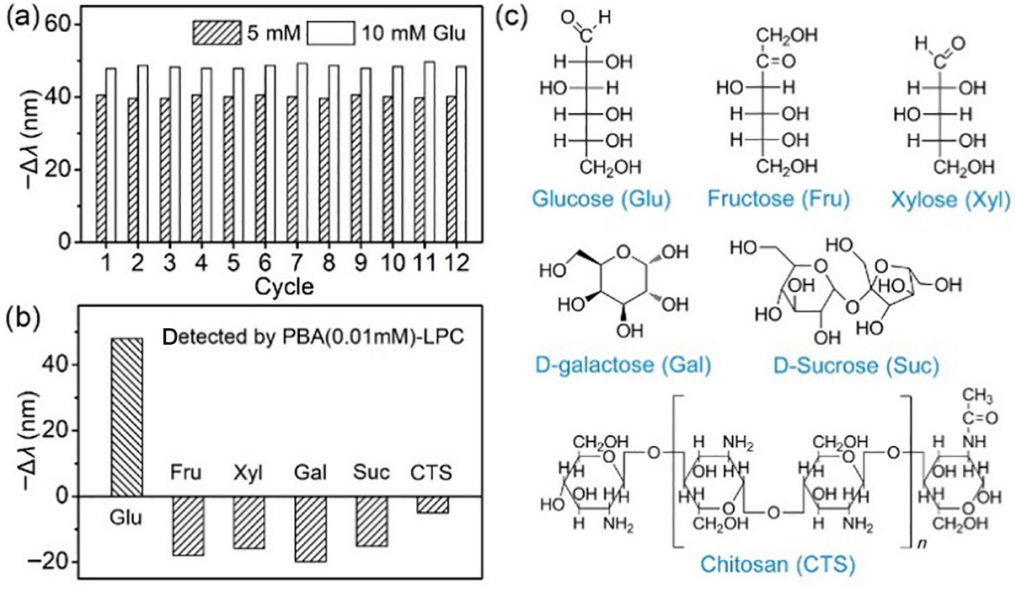

Figure 5 Reproducibility and selectivity for glucose detection. (a) The reflection blueshifts of the SiO_2/(PBA-EG) LPC reagents in twelve parallel tests of the glucose concentration (5 and 10 mM); (b) the PBA-LPC reagent selectively responded to the glucose in the way of reflection blueshift among all sugar tests; (c) the molecular structure for glucose, fructose, xylose, D-galactose, D-Sucrose, and chitosan. The concentration of all sugar solution was 10 mM except that of chitosan was 1 mM.

The interference by NaCl was first studied as it had a high concentration in the blood. Here, we prepared five SiO_2/(PBA-solvent) LPC reagents composed of EG, PCb, BDO, DMSO, and TEG with PBA dosage all set as 0.01 mM. Then, we compared their sensing performances for the detection of glucose solution (8 mM) under the interference of NaCl (Fig. 6 and Fig. S4 in the ESM). In the absence of NaCl in the glucose solution, the reflection peak of the mixture of PBA-LPC reagent and glucose solution blue-shifted 45 nm and its intensity decreased to 51%. While, in the presence of NaCl in the glucose solution, the reflection peak of the mixture blue-shifted 73 nm and its intensity decreased to 36%, accordingly. Through a group of parallel experiments, the reflection blueshift ($-\Delta\lambda$) increased with the increase of NaCl concentration, which indicated a higher response sensitivity. However, the reflection intensity of the mixture (R^*) which affected the detection reliability declined with the increase of NaCl concentration. It was necessary to choose a proper PBA-LPC reagent for the comprehensive consideration of sensitivity and reliability because a disappeared reflection at a high NaCl concentration directly led to the failure of detection. Among all the PBA-LPC reagents, the SiO_2/(PBA-PCb) reagent showed a detectable reflection at C_{NaCl} of 140 mM, which reached the NaCl concentration in the physiological environment (Fig. S5 in the ESM).

For the anti-interference capability to other chemical species, we also used the five PBA-LPC reagents to detect the same glucose solution and compared their sensing performances under the interference of $CaSO_4$, KH_2PO_4, and Vitamin C (Fig. 7 and Figs. S6–S8 in the ESM). Here, the reflection blueshift and the reflection intensity of the reagent-analyte mixture were compared in the absence or presence of $CaSO_4$, KH_2PO_4, and Vitamin C in the glucose solution. The concentrations of $CaSO_4$, KH_2PO_4, and Vitamin C were set as 2.5, 3.15 mM, and 10 μg/mL, which were their typical concentrations in the physiological environment. A close $\Delta\lambda$ and R in the absence or presence of distractors indicated a strong anti-interference capability for the PBA-LPC reagent. When the reflection intensity is adequately strong to determine the wavelength, a close $\Delta\lambda$ was the major criterion. The results suggested that most LPC reagents besides SiO_2/(PBA-TEG) reported an accurate response under the interference of $CaSO_4$. The SiO_2/(PBA-BDO) LPC reagent had the best accuracy under the interference of KH_2PO_4. All LPC reagents except for SiO_2/(PBA-DMSO) could be used under the interference of Vitamin C. Considering the response error in all cases, the SiO_2/(PBA-PCb) LPC reagent was the optimized detection reagents, which require fewer corrections to the working curve of "$\Delta\lambda$–C_{Glu}".

4 Conclusions

In summary, a PBA-functionalized SiO_2 liquid PC reagent was developed for convenient and reliable detection of glucose. When the PBA-LPC reagent was mixed with the analyte solution, the reflection peak blue shifted with the increase of glucose concentration because the esterification of boronic acid released H^+ ions, inhibited the deprotonation of the silanol group, weakened the particle surface charge, and eventually compressed the crystal lattice of LPC. Based on the above working mechanism, the glucose detection range amplified, the response sensitivity decreased, and the detection accuracy decreased with the increase of PBA dosage. The LPC reagent showed reproducible responses and reliable detection results in the parallel tests due to the precise introduction

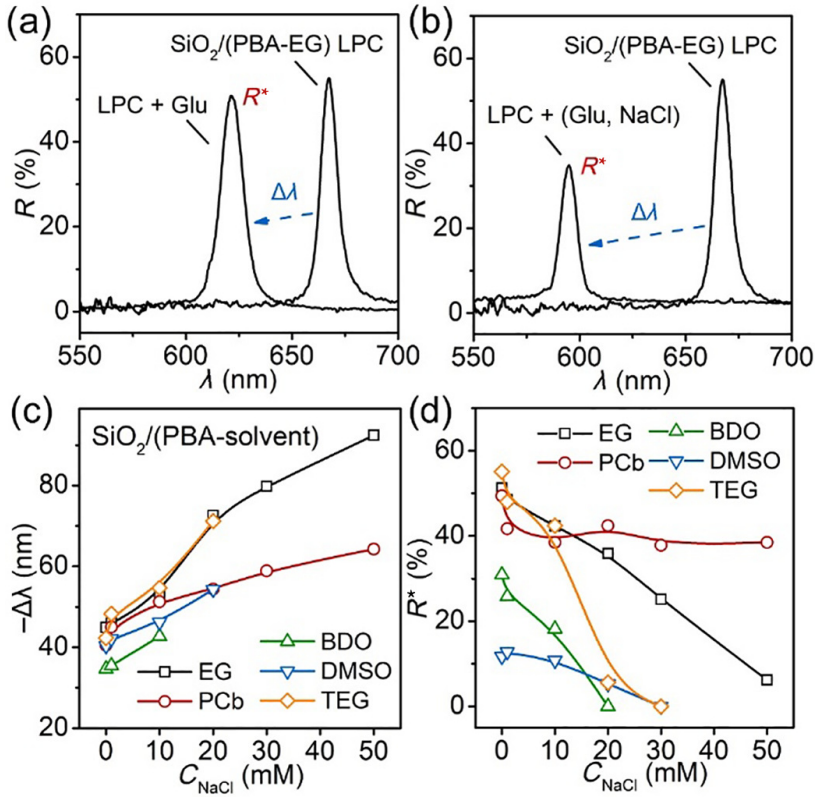

Figure 6 Interference by NaCl. (a) and (b) The optical response of the SiO_2/(PBA-EG) LPC to the glucose solution with and without the presence of 20-mM NaCl; (c) and (d) the influence of C_{NaCl} upon the reflection blueshift ($-\Delta\lambda$) and the reflection intensity after mixing (R^*). The PBA dosages in all SiO_2/(PBA-solvent) LPCs were 0.01 mM and the C_{Glu} was 8 mM for all tests.

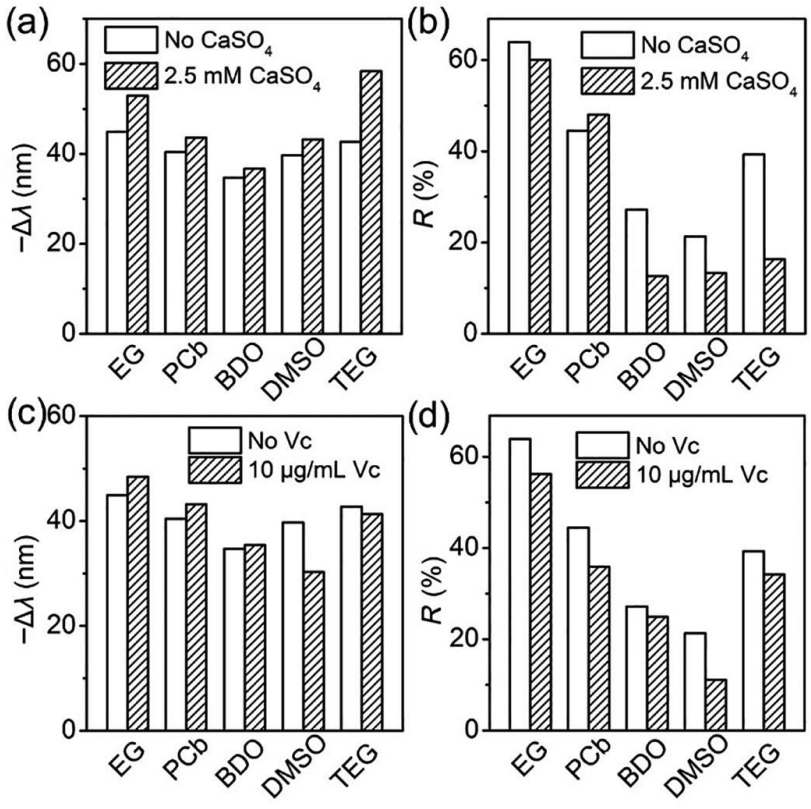

Figure 7 Anti-interference capability. The comparison of the reflection blueshift ($-\Delta\lambda$) and the reflection intensity after mixing (R) in the glucose detection with and without the interference of (a) and (b) $CaSO_4$ and (c) and (d) Vitamin C. The PBA dosages in all SiO_2/(PBA-solvent) LPCs were 0.01 mM and the C_{Glu} was 8 mM for all tests.

of the responsive substance (PBA) and its sufficient interaction with the analyte (glucose). It also showed a selective response to glucose which addressed the disturbance of other sugar molecules like fructose and galactose. The PBA-LPC reagent responded to glucose in a broad range, which covered the typical C_{glu} in the blood. Through the optimization of solvent in LPC, the SiO_2/(PBA-PCb) LPC reagent could function well in the presence of NaCl, $CaSO_4$, KH_2PO_4, and Vitamin C with typical concentrations in the blood. All these characteristics suggested its potential application for convenient blood sugar tests.

Electronic Supplementary Material: Supplementary material (influence of pH on detection; optical response of LPC with different PBA dosage to the glucose solution; comparison of glucose detection by LPC, PC gel, and imprinted PC sensors; glucose detection in the presence of NaCl, $CaSO_4$, KH_2PO_4, and Vc) is available in the online version of this article at https://doi.org/10.26599/NR.2025.94907166.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

Y. X. Z.: Data curation, validation, writing manuscript. H. M. Z.: Data curation, experimental design. J. P. G.: Project administration, funding acquisition, writing manuscript. All the authors have approved the final manuscript.

Use of AI statement

None.

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