



OPEN Plasmonic smart contact lens based on etalon nanostructure for tear glucose sensing

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Smart contact lenses, one of the most advanced wearable platforms, offer a combination of optical and electronic technologies that provide exceptional capabilities in various fields. These lenses are equipped with biosensors, microchips, and sometimes even miniature displays that enable the collection and analysis of biological and environmental data. Smart contact lenses represent a big leap in wearable technology and biosensors, potentially providing a new future for human interaction with the digital world, improving personal health, and promising a hopeful future in vision and wearable technologies. In this study, smart contact lenses based on plasmonic etalon nanostructure have been proposed and fabricated for tear glucose sensing. A cost-effective and simple technique of soft nanolithography has been proposed to fabricate a plasmonic sensor chip on a contact lens, enabling the detection of glucose solutions at different concentrations of 0.15, 1.5, 5, and 10 mM in a phosphate-buffered saline (PBS) solution. The proposed plasmonic etalon-based smart contact lens as a wearable platform exhibits the capacity to sense tear glucose (even at low concentration values) and offers relatively high sensitivity for non-invasive tear glucose sensing applications. The biocompatibility, cost-effectiveness, stability, and simple production of these contact lenses may provide new perspectives on wearable biosensors.

Keywords Smart contact lens, Eye wearable, Plasmonic, Biosensor, Tear glucose

In recent years, there has been a growing focus on improving the quality of human life, which has led to significant attention on physiological monitoring systems for disease diagnosis. A remarkable example of such a system is wearable biosensors^{1–8}, which are non-invasive and continuous platforms that enable the tracking of vital signs and metabolites from various parts of the body. The appeal of wearable biosensors lies in their ability to offer constant and real-time monitoring of biochemical indicators in biological fluids such as sweat, tears, saliva, and interstitial fluids through dynamic and noninvasive techniques. Also, wearable electronics are designed to be worn by individuals to continuously and precisely track their activities without interrupting or restricting the user's movements. Recently, wearable electronic devices that detect physiological changes to diagnose diseases have attracted widespread interest globally.

Among different anatomical structures, the eyes provide valuable physiological information and offer great potential as sensing sites for tracking biological signals. The composition of tear fluid is closely related to blood due to plasma leakage from blood to tear fluid through the blood-tear barrier, which separates the two fluids^{9–12}. Biomarker molecules in tears diffuse from the blood through the blood-tear barrier and strongly correlate with their concentration in the blood. Hence, the physical and chemical signals originating from the eye can be utilized to develop biosensors embedded in contact lenses. These molecules, which generate signals, are known as biomarkers and can have unique physical, chemical, or biological properties. For example, tear glucose concentration can be monitored to reflect blood glucose levels^{13–16}, while changes in intraocular pressure (IOP)^{17–21} can be monitored to reflect glaucoma severity.

Contact lenses, mostly utilized to correct vision and cosmetic purposes, serve as a unique wearable platform for tracking biological signals in the eye and managing diseases due to continuous contact with the tear fluid. These physical and chemical signals reflect the physiological state of the eye and the overall body, which offers a solution for early predicting and treating human eye diseases. Recently, plasmonic eye-wearables such as eyeglasses²² and contact lenses^{23,24} based on plasmonic nanostructures have attracted significant attention for color vision deficiency (CVD) management due to their outstanding properties and the adjustability of plasmonic

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resonances. Consequently, the potential of biosensors embedded within contact lenses for the monitoring and medical care of ocular disorders has attracted the interest of researchers^{14,25,26}.

In addition to smart contact lenses, eye-related wearable electronic devices such as smart glasses and artificial eyes are currently under development. Smart glasses are wearable computing devices equipped with electronic devices that can image or transmit information in the field of view²⁷. Furthermore, integrating electronics into soft contact lenses presents significant challenges due to the need for reliability during repeated eye blinking and flexibility, stretchability, and optical clarity for unobstructed vision. However, the most advanced contact lens sensors currently rely on opaque electronic components fabricated on plastic substrates with low oxygen permeability, rather than soft hydrogel contact lenses, which can limit the safe operation of the devices within the living eye^{28–32}. To address these challenges and develop a wearable sensor, it is necessary to utilize a commercial contact lens as the substrate while ensuring that the constituent materials of the sensor and antenna are transparent, stretchable, and biocompatible. Despite the considerable research efforts towards developing smart contact lenses, they are still in the early stages of performance as a smart wearable platform for monitoring various biophysical/chemical conditions. In today's world, the development of accessible tools for remote, user-friendly, and cost-effective diagnosis and treatment is of great importance.

Among the various biomarkers, glucose holds special significance in the field of diagnosis and treatment of diabetes. Diabetic individuals commonly use the finger prick approach daily to monitor their blood glucose levels, but this method is associated with pain and discomfort during sampling. Monitoring glucose levels in other bodily fluids like urine, saliva, intestinal fluid, or tear fluid could provide a non-invasive approach to diagnose and manage diabetes. Tear fluid has recently emerged as a promising physiological fluid for continuous glucose level monitoring through innovative sensor technologies. In addition to glucose, intraocular pressure serves as a crucial health indicator that can be assessed through the human eye. Elevated intraocular pressure stands as a major risk factor for glaucoma^{31,33–35}, a main cause of human blindness. Therefore, early diagnosis and management of glaucoma are essential, although challenging due to the gradual and asymptomatic nature of the disease.

In the present study, smart contact lenses based on plasmonic etalon nanostructure have been suggested and developed specifically for tear glucose sensing. A low-cost and simple technique using soft nanolithography has been proposed to fabricate a plasmonic sensor chip on a contact lens substrate. This sensor chip can detect different concentrations of glucose solutions (ranging from 0.15 to 10 mM) in phosphate-buffered saline (PBS). The proposed plasmonic smart lens as a wearable platform demonstrates the ability to sense tear glucose even at low concentration values and offers relatively good sensitivity for non-invasive tear glucose monitoring.

Experimental methods

Materials

Polydimethylsiloxane (SYLGARD 184 DOW CORNING, Sigma Aldrich), polyacrylamide (PAAM), and sodium alginate (SA) are utilized for fabricating contact lenses, while calcium chloride (CaCl_2) powder has been used as a cross-linking agent for polyacrylamide hydrogel. Phosphate-buffered saline (PBS) tablets have also been used to prepare a solution that simulates tear fluid.

Fabrication process

The plasmonic smart contact lens based on the etalon nanostructure was fabricated using polydimethylsiloxane (PDMS) polymer and polyacrylamide-sodium alginate (PAAM-SA) hydrogel through a cost-effective and simple soft nanolithography technique and investigated for its glucose-sensing capabilities.

Polyacrylamide-alginate hydrogels are frequently utilized in medical applications due to their biocompatibility, biodegradability, and tunable properties. It is essential to enhance its mechanical properties to fabricate a contact lens using this hydrogel. For this purpose, polydimethylsiloxane polymer, known for its biocompatibility, flexibility, and optical transparency, has been utilized to boost the mechanical properties of the hydrogel. This non-toxic and biocompatible material has gained popularity for different applications in the fields of biology^{36–41}, medicine^{42,43}, and Chemistry⁴⁴. Numerous studies have experimentally confirmed the superior flexibility and stretchability of PDMS^{45–47}. Its molecular structure, characterized by long siloxane chains (Si-O) with methyl groups, allows for significant deformation without breakage^{48,49}. PDMS can stretch up to 1000% before failure, indicating its remarkable flexibility, which is often measured through tensile tests⁵⁰. This material can endure substantial mechanical stress while maintaining its structural integrity and is capable of adapting to dynamic shapes without permanent deformation. Additionally, PDMS exhibits viscoelastic behavior, demonstrating both elastic and viscous properties, allowing it to return to its original form after being stretched⁴⁵.

Both polyacrylamide (PAAM) and polydimethylsiloxane (PDMS) are biocompatible polymers that are commonly utilized in biomedical applications. The combination of these two materials can yield a hydrogel characterized by improved biocompatibility and mechanical properties, making it suitable for tissue engineering, drug delivery, and other biomedical applications.

PDMS is known for its high flexibility, which can impart these properties to the PAAM hydrogel, leading to a hydrogel with improved flexibility and resistance to mechanical stress. Moreover, due to its superior tear resistance, PDMS can aid in preventing crack propagation and enhance the durability of PAAM hydrogel. By modifying the ratio of PAAM to PDMS in the hydrogel formulation, its mechanical properties can be adjusted to satisfy specific needs for different applications.

The schematic illustrating the fabrication process of plasmonic smart contact lenses based on PDMS-PAAM-SA polymer composite is shown in Fig. 1. First, homogeneous solutions of 1 volume percent of PAAM and alginate were prepared by dissolving appropriate amounts of polyacrylamide and sodium alginate powders in deionized water. Subsequently, polyacrylamide and alginate solutions were mixed with an equal ratio of 1:1 to

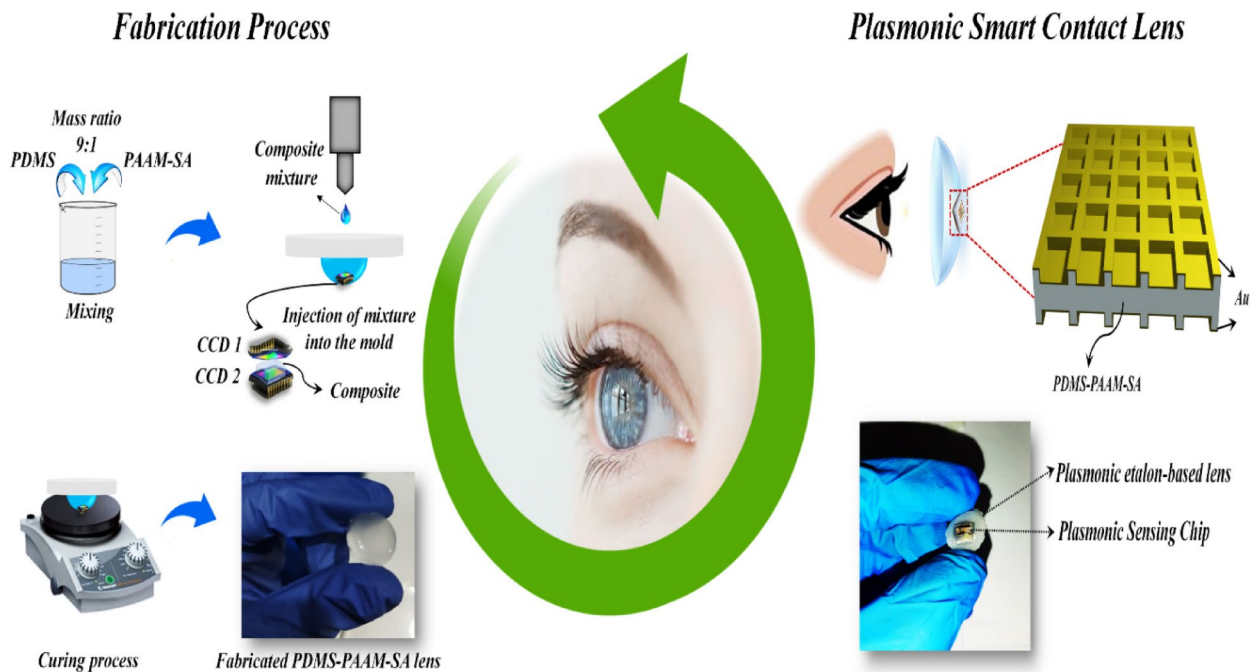


Fig. 1. A schematic of the manufacturing process of plasmonic smart contact lenses based on etalon nanostructure, real images of a simple lens based on PDMS-PAAM-SA polymer composite, and a plasmonic lens based on etalon nanostructure for glucose sensing were attached.

obtain 0.5% PAAM-SA solution. Next, a calcium chloride solution with a concentration of 200 mM was added to the PAAM-SA mixture to facilitate the formation of a hydrogel.

It should be noted that calcium chloride solution acts as a cross-linking agent for polyacrylamide hydrogel, forming ionic bonds with polymer chains and thus establishing a network structure. Higher concentrations of calcium chloride result in an enhanced degree of cross-linking and a more compact network. Also, sodium alginate acts as a gelling agent, facilitating the initiation of the cross-linking reaction and the formation of a stable hydrogel network. When alginate powder is mixed with acrylamide monomers and the CaCl_2 cross-linking agent, the gelation process takes place. This process involves the formation of three-dimensional networks of polymer chains that trap water molecules within the hydrogel structure.

Next, to prepare PDMS, a mixture of polydimethylsiloxane and curing agent was prepared with a weight ratio of 10:1, and these two components were combined and mixed for 5 min using a DC mixer to achieve a homogeneous mixture. Following this step, the combination of PAAM-SA hydrogel and PDMS at a weight ratio of 1:9 was mixed with a DC stirrer and placed in a vacuum chamber for 10 min to remove bubbles. This process resulted in forming a homogeneous polymer composite PDMS-PAAM-SA, which is suitable for contact lens manufacturing.

Contact lenses must be curved due to the natural curvature of the cornea, and conventional lithography techniques can only be used on flat surfaces. This study introduces a simple and low-cost technique utilizing soft nanolithography to fabricate a plasmonic etalon nanostructure on the curved surface of the lens. In this process, the charge-coupled device (CCD) of the camera, which has a two-dimensional periodic square pattern with excellent resolution, is used as a stamp.

To initiate, the first CCD is placed in the lens mold and a mixture of PDMS-PAAM-SA is poured on it (Fig. 1). Subsequently, the second CCD is carefully placed onto the composite and aligned in front of the first CCD, with the curing process involving a gradual increase in temperature from 50 to 100 °C over 1 h. After 24 h, the biocompatible contact lens based on the etalon nanostructure was removed from the mold and the CCDs were carefully detached from the lens. Finally, a biocompatible double 2D grating (etalon) contact lens based on PDMS-PAAM-SA will be achieved, with a thin layer of 15 nm gold coated on both patterned surfaces of the lens using a sputter deposition device. In this manner, a flexible and biocompatible plasmonic lens was fabricated using a cost-effective and simple method (Fig. 1), compared to other expensive and complicated techniques, such as electron beam lithography, and suggested for eye tear glucose sensing.

Results and discussion

The fabricated plasmonic etalon (Fabry–Perot) is an optical structure with two parallel reflective surfaces made from plasmonic materials that support surface plasmon polaritons (SPPs), and enhance light-matter interaction. Its performance relies on light interference, where light reflects between the surfaces, creating constructive and destructive interference that results in distinct transmission peaks.

The metal layers of the plasmonic etalon play a crucial role by supporting surface plasmon resonances. As light interacts with these layers, a portion of its energy couples into surface plasmons, thereby enhancing the

electric field at the surface. This enhancement increases the device's sensitivity. The condition for constructive interference is given by $2nd = m\lambda$, where n is the refractive index, d is the distance between layers, m is the resonance order (an integer number), and λ is the wavelength of light in vacuum^{51,52}. The quality factor (Q-factor) of the etalon is critical as it determines the sharpness of the resonance peaks^{53,54}; higher Q-factors correlate with sharper peaks and greater sensitivity to variations in wavelength or refractive index.

When light interacts with the etalon structure, some is reflected while some transmits, influenced by the interference pattern from multiple reflections. The etalon can selectively transmit wavelengths based on its design, and this selectivity can be tuned by adjusting the angle of incidence or the refractive index of the medium between the reflective layers. So, plasmonic etalons are particularly sensitive to refractive index changes near their surfaces, making them effective for biosensing applications.

Since contact lenses interact directly with the eyes, it is essential that they possess good transparency alongside biocompatibility and do not hinder the individual's vision. As shown in the diagram of Fig. 2a, the proposed lenses based on the PDMS-PAAM-SA composite exhibit good transparency in the visible wavelength range.

The scanning electron microscope (SEM) image of the etalon-based plasmonic smart contact lens was recorded and shown in Fig. 2b. As can be seen, the proposed plasmonic smart lens has a high-resolution 2D periodic square pattern. Also, the schematic of the proposed smart lens structure based on plasmonic etalon is shown in Fig. 2c, which shows the arrangement of two-dimensional square periodic patterns on both sides of the structure.

Furthermore, the transmission spectrum of the proposed plasmonic contact lens was measured and depicted in Fig. 2d. The plasmonic resonance response of gold nanowires occurred at a wavelength of 560 nm, which corresponds to the surface plasmon lattice resonance (SLR) induced by the plasmonic etalon array on the lens. The arrangement of metal nanoparticles in a periodic lattice can exhibit highly narrow and intense resonances referred to as plasmonic surface lattice resonance (SLR). This phenomenon arises from the coupling between the diffracted waves (DO) in the periodic structure and the localized surface plasmon resonances (LSPRs) caused by the metal nanowires incorporated within the structure. The proposed plasmonic smart lens consists of a two-dimensional periodic array of gold nanowires, which can support diffraction order waves with low spectral width and LSPR modes.

It should be noted that the lattice constant (periodicity) of the structure is on the order of microns and due to the large dimensions of the unit cell, a direct matching between the wavelength of incident light in the visible region and the lattice constant (or periodicity) does not occur. However, the plasmonic surface lattice resonance has been detected in the experimental transmission spectrum (Fig. 2d). This physical phenomenon arises from the coupling between the local surface plasmon resonances (LSPRs) at the corners of each unit cell and the diffraction order waves within the two-dimensional periodic structure. Further, the distance between the neighboring unit cells for these CCD stamps is approximately 50–500 nm, and each side of the square unit cell acts as a rod that is coupled to the corresponding rod of the adjacent unit cell. In this way, each unit cell is coupled to the adjacent unit cell, thereby satisfying the condition of surface lattice resonance in the visible region for the proposed plasmonic structure.

In addition to the experimental and manufacturing aspects, the proposed plasmonic contact lens based on etalon nanostructure has been simulated using the Finite Difference Time Domain (FDTD) and Mode-solution modules of the Lumerical software. The dimensions and specifications of the structures are inputted into the software according to their experimental dimensions; the thickness of the gold thin layer is set at 15 nm and the mesh size in the x, y, and z directions is assumed to be 1.5 nm. In addition, the refractive index values of gold and PDMS-PAAM-SA composite were taken from the data presented by Johnson and Christy et al.⁵⁵ and the experimental data measured with a refractometer, respectively.

The transmission spectrum of the etalon-based plasmonic smart lens is simulated and presented in Fig. 2e; it is evident that a good agreement between the experimental and simulation results has been achieved and the resonant response of gold nanowires and modes of the etalon structure have appeared. For a more comprehensive analysis, the electric field distribution at the resonant wavelength is also simulated; the refractive index profile of the simulated plasmonic etalon structure and the electric field distribution at the resonance wavelength of 560 nm are given in the appendix of Fig. 2e. The simulated structure consists of 3×3 arrays of unit cells situated on both the top and bottom sides of the structure with a lattice constant of 2500 nm, the dimensions of which are confirmed by scanning electron microscope (SEM) images. As can be seen, the electric field localization in the etalon structure occurred at the resonance wavelength, which is caused by the lattice resonance of plasmonic gold nanowires in the etalon array.

In this research, the design of the etalon nanostructure on the curved surface of the lens for tear glucose sensing has been proposed, which provides new insights into wearable sensor technology. For this purpose, glucose solutions with different concentrations of 0.15, 1.5, 5, and 10 mM were prepared based on phosphate buffer salt solution (PBS). It should be noted that PBS solution was utilized to simulate tear fluid, each PBS tablet dissolved in 100 ml of deionized water containing sodium chloride (137 mM), potassium chloride (2.7 mM), and phosphate buffer (10 mM).

After immersing the plasmonic etalon-based smart lens into glucose solutions with different concentrations, the transmission spectrum of the lens was recorded (Fig. 3a–b). The results revealed that the depth of resonance increased with the increasing glucose concentration. Therefore, a logical trend emerges in the results of glucose sensing, with a good separation between optical responses as the concentration changes. The proposed plasmonic lens demonstrated its ability to respond even at low glucose concentrations (0.15 mM). The smart lens sensor chip exhibited excellent linear behavior in the concentration range of 0.15–5 mM (Fig. 3d), while the saturation effect occurred when the concentration increased to 10 mM (Fig. 3c). Since the concentration of tear glucose in healthy people is in the range of 0.16–0.5 mM and about 0.9 mM for those with diabetes, the proposed smart lens sensor chip has covered an appropriate range and offers the potential to function as a wearable platform capable

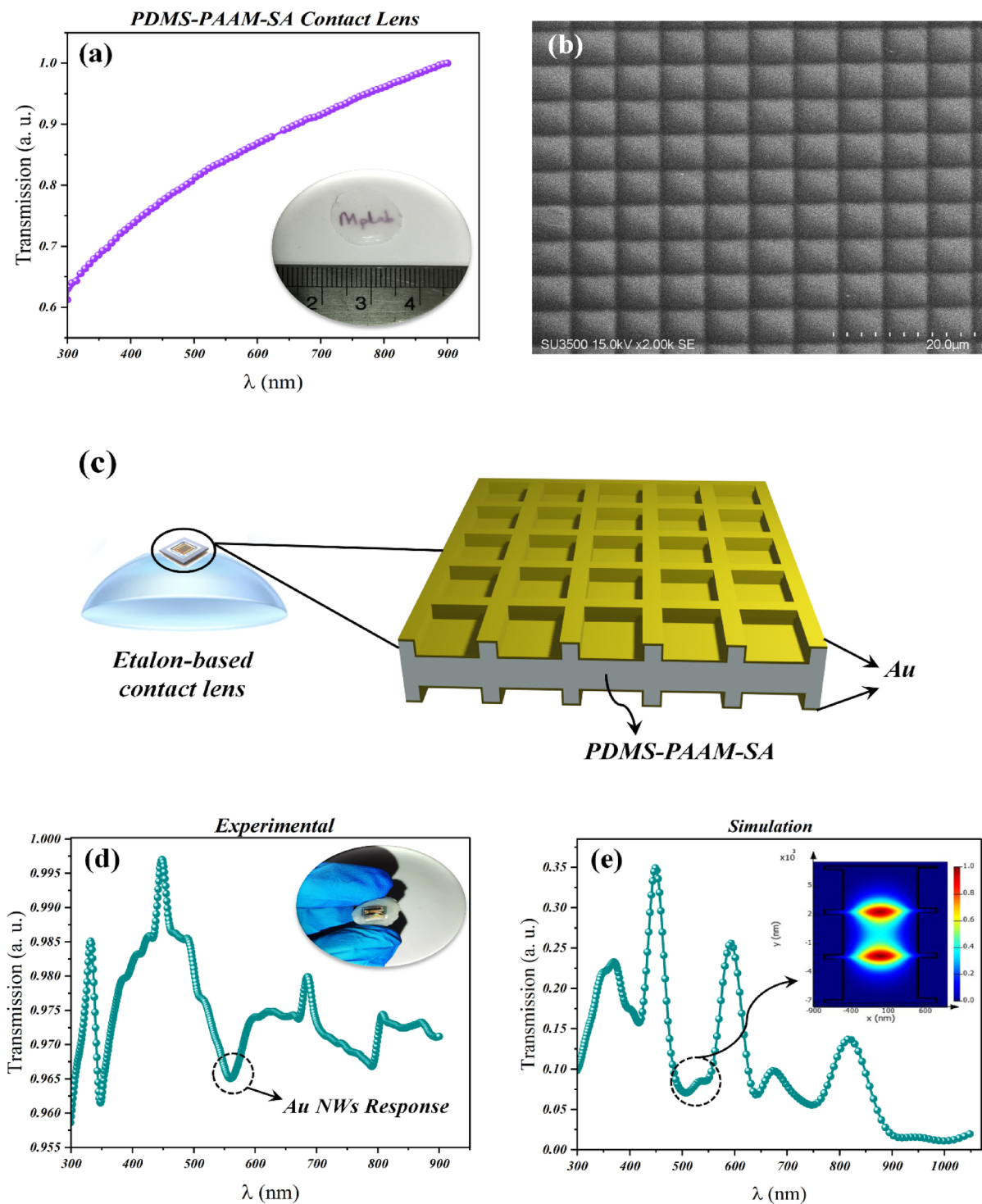


Fig. 2. (a) Transmission spectrum of a produced simple lens based on PDMS-PAAM-SA composite, the real image of the produced lens is attached, (b) the recorded SEM image of a plasmonic smart contact lens based on the etalon nanostructure, (c) Schematic of the fabricated plasmonic smart lens, (d) experimental and (e) simulated transmission spectrum of the plasmonic etalon-based lens. The electric field distribution of the etalon nanostructure at the resonance wavelength of 560 nm is given in the appendix of Fig. 2e.

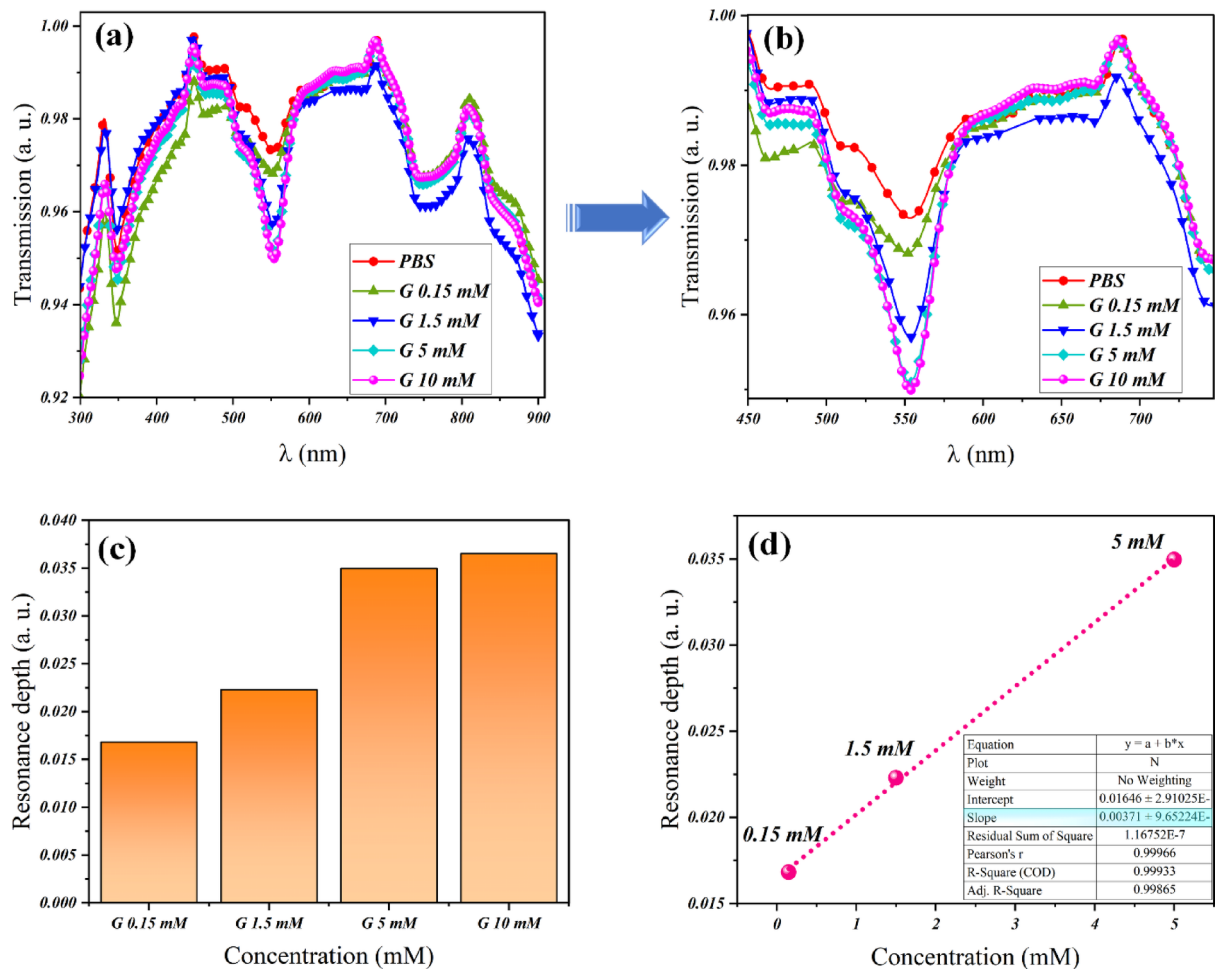


Fig. 3. (a) Experimental results of glucose sensing utilizing a plasmonic smart lens based on an etalon sensor chip for different glucose concentrations, (b) a magnified illustration of (a), and (c–d) the sensitivity of sensing for different glucose levels using the plasmonic etalon-based smart contact lens.

of detecting tear glucose, even at low concentration levels. The ability of the proposed plasmonic smart lens to sense different concentrations of tear glucose is directly proportional to the sensitivity ($S = 0.00371$ a. u./mM) of that sample (Fig. 3d), offering relatively good sensitivity for non-invasive tear glucose sensing applications.

As contact lenses come into direct contact with the eye, it is crucial to ensure their safety and compatibility with biological tissues. For this purpose, in vitro cytotoxicity test of the proposed plasmonic contact lenses was carried out according to ISO 10,993-5 standard method. The testing procedure was outlined in Table 1.

The results of the in vitro cytotoxicity assessment using L929 mouse fibroblast NCTC clone 929 strain L (as presented in Fig. 4 and detailed in Table 1) demonstrate that the proposed plasmonic contact lenses are non-toxic, with an average cell viability of 99.39%.

Conclusion

Eyes can provide rich physiological information and offer extensive diagnostic potential as a sensing site, making contact lens sensors applicable for the non-invasive monitoring of many diseases and conditions. Smart contact lenses, due to their multifunctional biosensor capabilities and highly integrated functionality, provide an excellent platform for eye disease diagnosis and drug delivery. In this study, plasmonic smart contact lenses based on etalon nanostructure have been proposed to assess glucose levels in tears. These lenses are fabricated through the soft nanolithography technique and can detect glucose in tears with remarkable accuracy and appropriate sensitivity. This sensor platform is suitable as a non-invasive method for measuring glucose in tears, particularly at low concentrations, and provides new possibilities for medical and research applications. The biocompatibility, low cost, stability, and simple manufacturing of these contact lenses can offer new insights into the potential applications of wearable biosensors.

Repeat/sample	Optical density (570 nm)	Acceptable limit		Results		
		Viability %	Morphological grade	Morphological grade	Viability %	Average viability %
TEST SAMPLE 1	0.883	> 70%	< 2	0	100.46	99.39
TEST SAMPLE 2	0.926			0	99.89	
TEST SAMPLE 3	0.896			0	98.14	
TEST SAMPLE 4	0.901			0	101.12	
TEST SAMPLE 5	0.884			0	97.36	
CONTROL SAMPLE 1	0.879	100%	0	0	100.00	100
CONTROL SAMPLE 2	0.927			0	100.00	
CONTROL SAMPLE 3	0.913			0	100.00	
CONTROL SAMPLE 4	0.891			0	100.00	
CONTROL SAMPLE 5	0.908			0	100.00	
Total Average of test sample				0	99.39	

Table 1. Qualitative morphological grading and quantitative measurements of cytotoxic effects by MTT assay after 24 h.

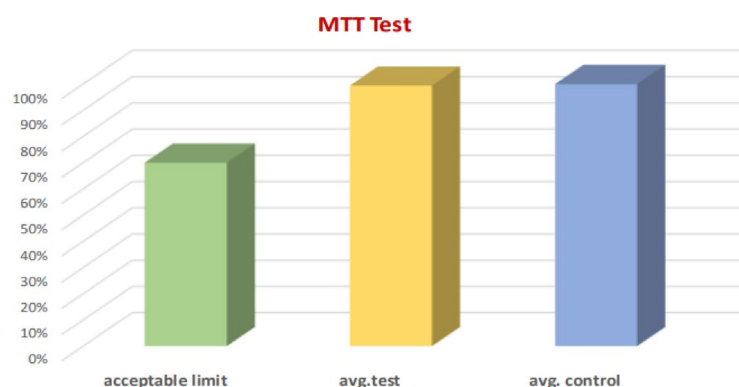


Fig. 4. Results of the MTT test for the proposed plasmonic smart contact lens.

Data availability

All of data will be available by request from corresponding author.

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Author contributions

N. Roosatei do the experimental work and write the main text and S. M. Hamidi supervised the work and edit the main text.

Declarations

Competing interests

The authors declare no competing interests.

Additional information

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