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Mini review

Antifouling Self-Assembled Monolayers for Designing of Electrochemical Biosensors

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Antifouling sensing interfaces can reduce or eliminate the non-specific absorption in electrochemical biosensing. Many antifouling strategies have been reported in the references. Self-assembled monolayer (SAM) is inexpensive and versatile for immobilization of biorecognition element onto the surface of transducer. In this work, we highlight the recent progress in antifouling SAMs and addresed their applications in electrochemical biosensors, including phenyl phosphates, small molecules, peptides and PEGylated conductive films.

Keywords: antifouling; electrochemical biosensors; self-assembled monolayer

1. INTRODUCTION

Electrochemical biosensors have received prominence in the detection of biological molecules or even whole cells [1]. However, because numberous proteins existing in bodily fluids may non-specifically adsorb onto the sensing interface, electrochemical biosensors rise to the challenge for the detection of target proteins in complex biological samples with high sensitivity, specificity and reproducibility [2-4]. Therefore, a nonfouling interface is required to resist the non-specific adsorption from proteins.

Self-assembled monolayer (SAM), in particular alkanethiol SAM, is inexpensive and versatile for immobilization of biorecognition element onto the surface of transducer [5, 6]. Usually, the long-chain SAM chain can enhance the stability and efficiency of the sensing interface in reducing/eliminating the non-specific absorption. For example, the SAMs of 11-mercaptoundecanoic

acid (MUA), poly(ethylene glycol) (PEG) and oligo(ethylene glycol) (OEG) have shown excellent antifouling properties [7-9]. However, the long-chain SAM is limited in electrochemical analysis because it causes a low permeability to electron transfer [10]. This disadvantage could be circumvented by employing a short-chain or alkanethiol mixed SAM. However, such an interface exhibits low biomolecule immobilization capacity and increase the non-specific interactions, thus negatively influencing the performances of electrochemical biosensor. To reduce the non-specific adsorption of interferences on the peptide-modified electrode surface, usually, a commonly used strategy is to block the nonsensing regions of biointerface with a medium length thiol molecule, such as 1-hexanethiol (HT) or 6-mercapto-1-hexanol (MCH) [11-14]. The six-carbon SAM allows for the electron transfer between the electrode and the electroactive species, but requires two-step procedures for the preparation of sensing surface and exhibits poor capability in resistance of non-specific protein adsorption against the undiluted serum samples. Recently, novel SAMs including phenyl phosphates, small molecules, peptides and PEGylated films have been constructed on electrode surface to develop electrochemical biosensors. This paper summaried the progress in the reported SAMs and addresed their applications in electrochemical assays.

2. ANTIFOULING SAMS

2.1 Phenyl phosphates

Amphoteric polymers contain many positive and negative groups, such as carboxybetaine, sulfobetaine and phosphorylcholine (PC). For charged surfaces, the ionic strength and potentials play important roles in surface properties. Antifouling materials (e.g. polyethylene glycol and amphoteric polymer) can prevent non-specific protein adsorption and cell adhesion, thus having been used as the electrode modifiers in biomedicine. Chen et al. studied the influence of ion and surface potential on the antifouling properties of poly(EDOT-PC) and poly(EDOT-EG) films [15]. The smooth poly(EDOT-PC) and poly(EDOT-EG) films were successfully prepared by a simple surface coating method for the first time. The results indicated that the two films can effectively adsorb anions and enhance their hydration. The synergistic effect of ion and surface potential on the antifouling performance of poly (3,4-ethylenedioxythiophene) was then compared with that of the polyethylene glycol and phosphorylcholine. Jiang et al. prepared a mixed SAM composed of amphoteric phenyl phosphorylcholine (PPC) and phenylbutyric acid (PBA) (Figure 1) [16]. PPC was responsible for inhibiting the non-specific adsorption of proteins and PBA was used as the biological recognition element to capture the antibody. The results demonstrated that the sensor showed specific recognition ability for tumor necrosis factor (TNF-α) in whole blood. The linear detection range is 0.01 ~ 500 ng/mL with a detection limit of 10 pg/mL. The result is comparable to that achieved by the commercial enzyme-linked immunosorbent assay (ELISA) kit, suggesting that the sensor exhibits great potential for clinical applications.

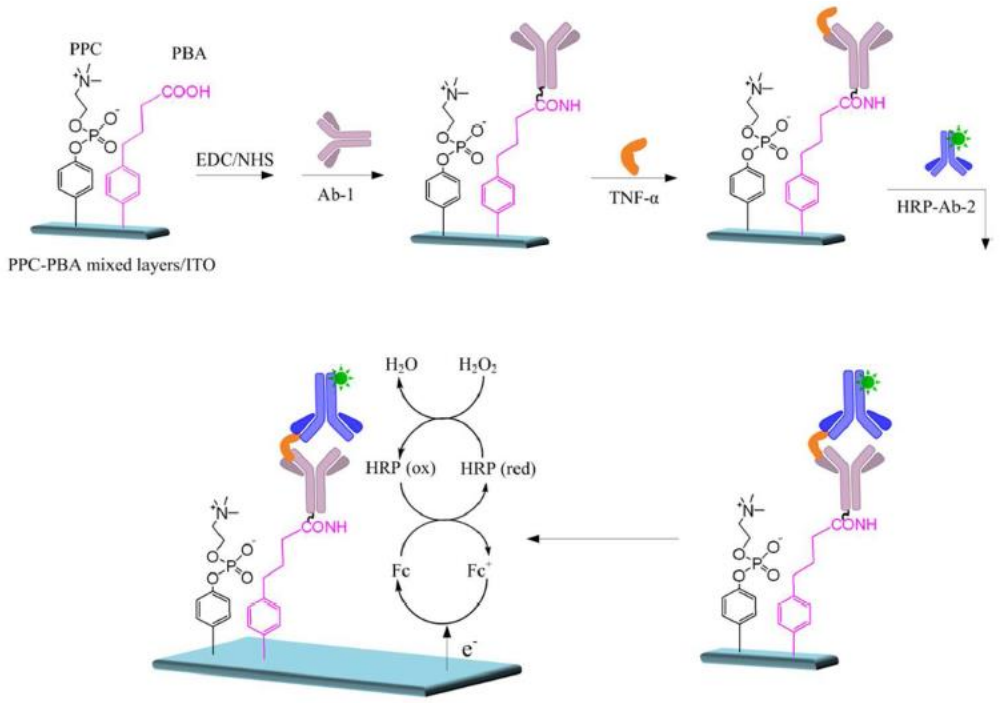

Figure 1. Illustration of the PPC-PBA mixed layers/ITO based immunosensor for the detection of TNF-α. Reprinted with permission from reference [16]. Copyright 2016 American Chemical Society.

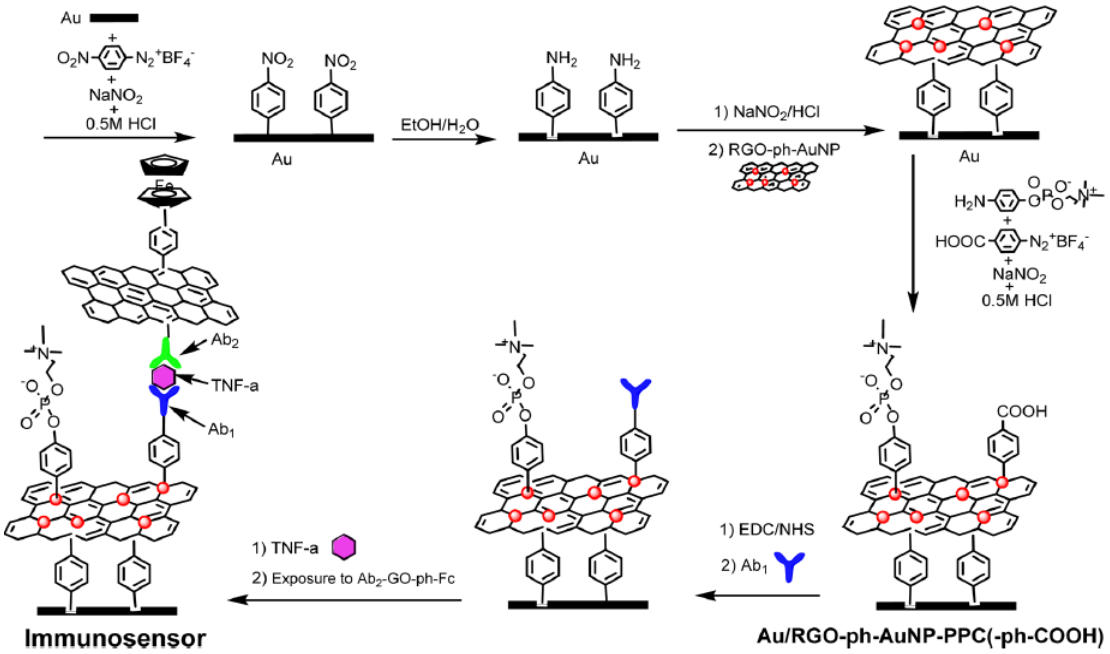

Figure 2. Schematic of the fabricated immunosensor for detection of TNF-α. Reprinted with permission from reference [17]. Copyright 2016 American Chemical Society.

Graphene oxide (GO) and gold nanoparticles (AuNPs) have excellent propensities for electrochemical signal amplification, and have widely applied in the fabrication of electrochemical sensing devices. Qi et al. developed a sandwich-like electrochemical immunosensor for the detection

of TNF-α (Figure 2) [17]. Firstly, the Au-RGO-ph-AuNP-ph-PPC(-ph-COOH) with antifouling property was modified on the electrode surface for the immobilization of anti-TNF-α. The signal was recorded by monitoring the oxidation of ferrocene (Fc) tag in the Ab_2-GO-ph-Fc conjugate. The detection range of this method is 0.1 ~ 150 pg/mL, and the detection limit is 0.1 pg/mL. The immunosensor has good antifouling performance, high sensitivity and specificity. At the same time, a mixed layer of 4-carbxyphenyl and 4-aminophenyl phosphorylcholine amphoteric molecule has good biological inhibition effect without causing high impedance. This proof of concept has an excellent application prospect in designing of biosensors for clinical applications.

2.2 Small molecules

The formation of SAM on the electrode surface is quick by the covalent interaction between mercaptan and metal surface. Fragoso et al. reported a novel sulfydryl SAM composed of 3,5-dihydroxybenzyl alcohol containing carboxyl and hydroxyl end groups. The SAM-covered electrode was used to develop a label-free biosensor for detection of prostate specific antigen (PSA) by electrochemical impedance spectroscopy (EIS). The detection limit as low as 9 ng/mL was achieved [18]. Zaitouna et al. reported three sulfydryl antifouling agents and investigated their antifouling ablility for the detection of HIV, including methoxy-terminated diluent (C6-MEG), mannose diluents (C6-MAN) and ethylene-glycol-terminated diluents (C6-EG) [19]. Among them, C6-MEG exhibited the highest antifouling ablility to prevent non-specific adsorption for real sample assays. Liu et al. reported a sandwich-like electrochemical biosensor with high sensitivity for lysozyme detection based on the signal amplification of horseradish peroxidase (HRP) [20]. The antifouling interface of the sensor consists of Lys binding aptamer (LBA), dithiothreitol (DTT) and mercaptohexanol (MCH). Lys was modified on the interface by the interaction with LBA, and AuNP functionalized with HRP and LBA (HRP-AuNP-LBA) was connected with Lys to form a sandwich structure. The signal was collected by differential pulse voltvammetry (DPV). The linear detection range is 0.01 ~1×10^5 pg/mL with a detection limit of 0.003 pg/mL. Zhang et al. reported an ultrasensitive biosensor for ATP detection using polydopamine (PDA)/reduced graphene oxide carbon nanofiber (GO-CNF) (PDA/GO-CNF)-modified electrode [21]. Cysteine (Cys) was used as the antifouling agent. This aptasensor has a good linear range for ATP detection (0.1 pM ~ 5 nM) and a low detection limit (13 fM).

Bovine serum albumin (BSA) has relatively large size and molecular weight (~66500 Da), which shows high steric hindrance effect for the antigen-antibody interaction. Thus, BSA-modified electrode is more suitable for analysis of small molecules. It has been suggested that a small inactive peptide, oxidized glutathione (GSSG), can be used instead of BSA as a more efficient blocking agent for immunosensor (Figure 3) [22]. GSSG was much smaller than the antigen such as EGF, which did not hinder the antigen-antibody interaction with improving sensitivity. The linear range is 0.1 pM ~ 0.1 μM for EGF detection, and the detection limit is 0.01 pM.

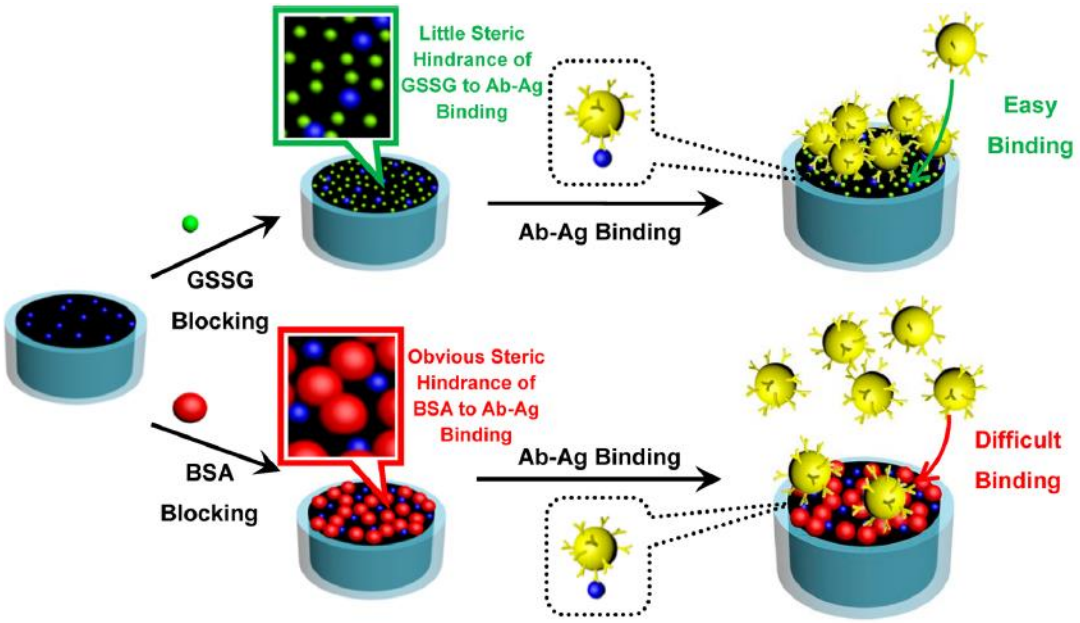

Figure 3. Schematic representation of two different blocking processes using GSSG and BSA as blocking reagent to design the electrochemical immunosensor, respectively. Reprinted with permission from reference [22]. Copyright 2015 American Chemical Society.

2.3 Peptides

Peptides play critical roles in the regulation of numerous biological processes, such as enzyme activation, intermediary metabolism, membrane permeability and protein synthesis [23, 24]. Changes in the levels of peptides may be indicated of disease progression, bacterial infection, hormone level and so on [25]. Thus, peptides have been regarded as the biomarkers for the early detection of many diseases. Moreover, peptide can be designed and applied as the biological recognition element to bind the target protein with a high specificity and a strong binding affinity [26-28]. Charged amino acid residues such as glutamic acid (E) and lysine (K) in the peptide sequence may produce electrostatic interaction among peptides, thus eliminating the non-specific adsorption. Nowinski et al. studied the effect of terminal amino acid residuces on the function of peptide, such as EKEKEKE-PPPPC-Am and EKEKEKE-C-Am (Figure 4) [29]. The two peptides have strong adsorption interaction for gold electrode, but the EKEKEKE-PPPPC-Am can form ordered secondary structure and has good antifouling ability for protein adsorption. In addition, the PPPPC segment has better binding effect than other segments, such as PC, PPC, and PPPC. The specific and non-specific adsorption of EKEKE-PPPPC-Am to cells was also investigated. Kakegawa et al. designed a oligopeptide of CGGGKEKEKEK in which the terminal cysteine residue can facilitate the immobilization of peptide [30]. The SAM of CGGGKEKEKEK on the electrode surface effectively enhanced the antifouling performance of the electrode. On this basis, RGD sequence for cell adhesion was included in the peptide for the recognition of target cells.

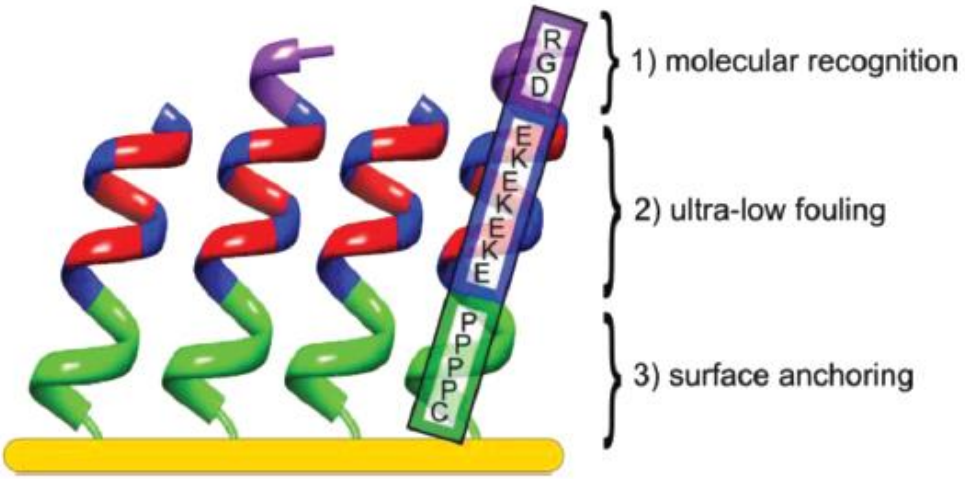

Figure 4. All-in-one natural peptide SAM on gold with three distinct functions incorporated into the peptide sequence. Reprinted with permission from reference [29]. Copyright 2012 American Chemical Society.

Using amphoteric peptide as the antifouling material, various electrochemical biosensors have been developed. The specificity and selectivity of electrochemical peptide biosensors have been improved by adding additional amino acids to the peptide recognition probes. For example, Zaitouna, et al. added an amino acid sequence (AA) to the N-terminal of thiolated methylene blue-modified peptide and studied its effect on the specific and selective detection of HIV [31]. It was found that the peptides exhibited distinctive properties and detection limits. Aptasensors are widely used in analytical chemistry and biochemistry because of their low cost, high selectivity, good stability and reversibility. However, in clinical analysis, especially in human serum solution and cell lysate, the non-specific adsorption will result in false-positive reaction, thus reducing the clinical applicability and sensitivity of aptasensors. AFP is a biomarker of liver cancer. Cui et al. designed an amphoteric polypeptide (EESKSESKSGGGGC) to effectively block the specific adsorption of other proteins (Figure 5) [32]. This allowed for the sensitive and specific detection of AFP with high pollution prevention. Cui et al. also developed a label-free electrochemical DNA biosensor for the detection of breast cancer susceptibility gene (BRCA1) based on the self-assembled antifouling peptide layer [33]. The linear detection range of this biosensor is 1.0 fM ~ 10.0 pM with a detection limit of 0.3 fM. Luo's group developed an antifouling and ultrasensitive electrochemical DNA biosensor by combining the amphoteric peptide of EKEKE with the conducting polymer poly (3,4-ethylenedioxythiophene) (PEDOT) [34]. The detection limit of this method is as low as 0.03 fM. The biosensor has strong antifouling ability, high sensitivity, good selectivity and high efficiency for detection of BRCA1 in real samples.

Liu et al. designed an amphoteric peptide of CPPPPEK2)/(EK)4(EK) (Figure 6) [35], in which the cysteine faciliated the immobilization of peptide through the Au-S interaction, and the PANI film on the electrode surface was used to hold the peptide upright. The steric repulsion provided by the branched peptide is beneficial to linear sequence correlation due to its high hydration and relative conformational entropy. MCF-7 cells (human breast cancer cell line) in blood were selectively captured by the redox PANI.

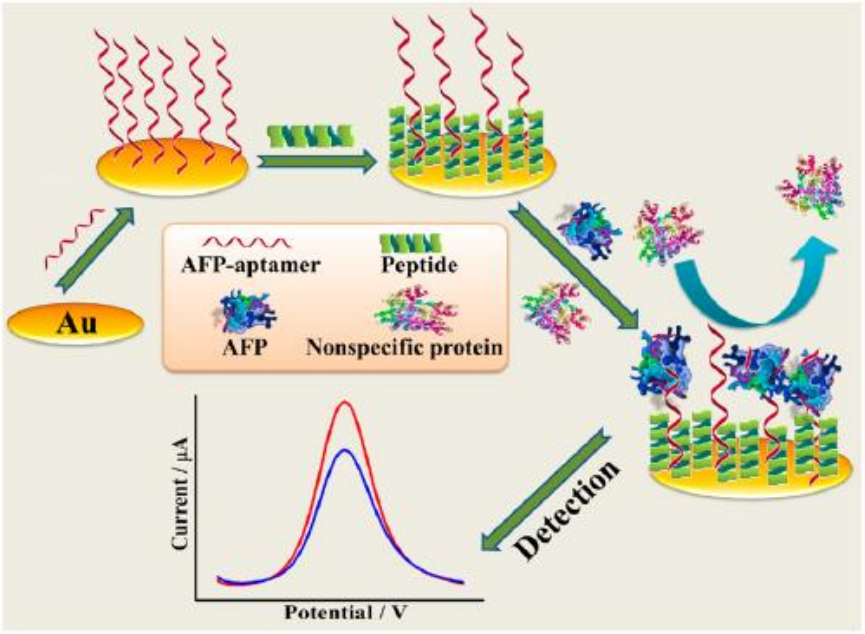

Figure 5. Schematic illustration of the AFP aptasensor fabrication protocol through the stepwise immobilizations of long-chained AFP-specific aptamers and short-chained peptides onto gold electrode surface via self-assembly. Reprinted with permission from reference [32]. Copyright 2017 American Chemical Society.

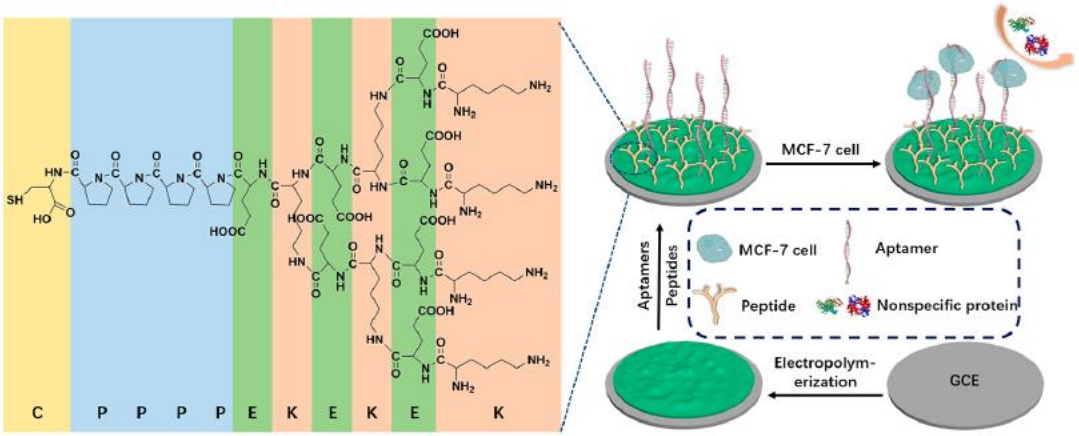

Figure 6. Schematic illustration of the preparation of a PANI-supported branched peptide-based amperometric cell sensor. Reprinted with permission from reference [35]. Copyright 2019 American Chemical Society.

Based on the antifouling peptide loaded with internal standard and electrochemical probes, a dual-mode electrochemical array was constructed to detect prostate specific antigen (PSA) (Figure 7) [36]. The two antifouling peptides were labeled with graphene oxide-Fe_3O_4-thionine (GO-Fe_3O_4-Thi) probe and internal reference ferrocene, respectively. The peptide-GO-Fe_3O_4-Thi probe contains a sequence of HSSKLQK which can be recognized and cleaved by PSA. This dual-mode antifouling electrochemical biosensor showed a wide linear range from 5 pg/mL to 10 ng/mL.

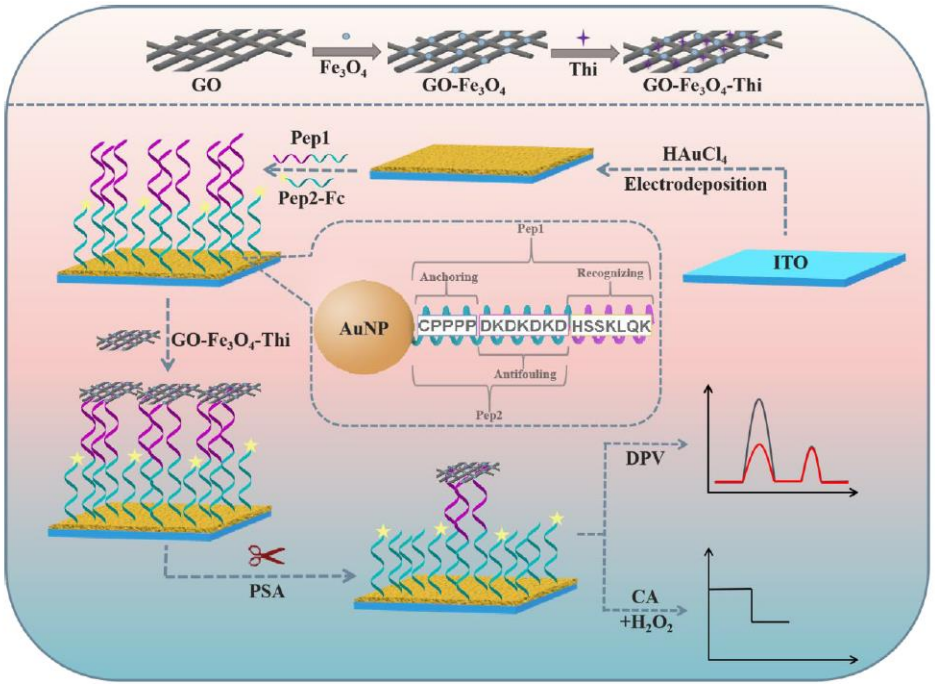

Figure 7. Schematic illustration of antifouling electrochemical biosensor working principle. Reprinted with permission from reference [36]. Copyright 2019 American Chemical Society.

Despite these achievements, it is still a difficult challenge to use a highly selective biosensor which can detect specific targets in complex biological samples. Ye et al. investigated the difference in antifouling ability between CRERERE and CYSYSYS for electrochemical detection [37]. The results of SPR analysis showed that the two peptide SAMs had an adsorption capacity in the range of 1.97-11.78 ng/cm^2 for a single protein. However, zwitterions have better antifouling effect for the detection of a single protein or comlicated protein samples. This result reveals the relationship between the charge of peptide and its antifouling function, and is of significance for the development of new antifouling peptide materials.

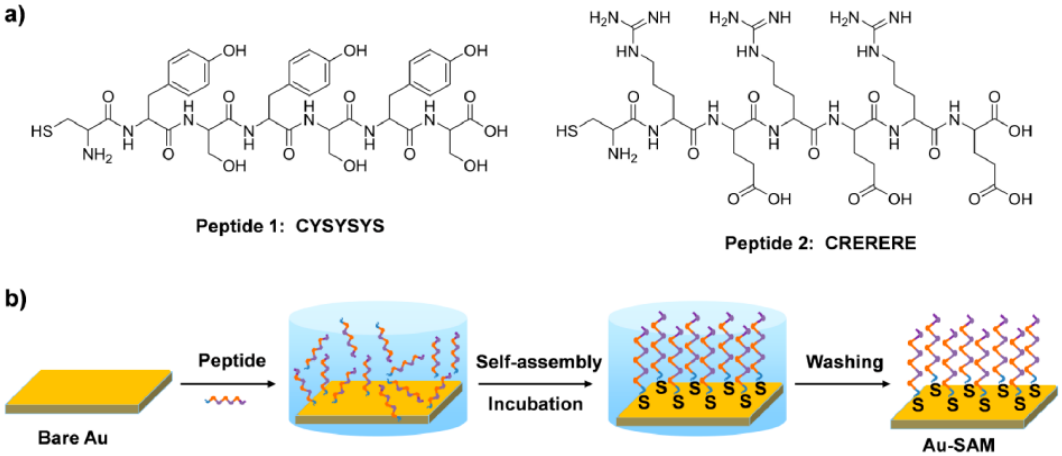

Figure 8. (a) Molecular structures of an amphiphilic peptide with a sequence of CYSYSYS and a zwitterionic peptide with a sequence of CRERERE and (b) achematic illustration of the attachment of peptide self-assembled monolayers (SAMs) on the Au Surfaces. Reprinted with permission from reference [37]. Copyright 2015 American Chemical Society.

Moreover, Liu et al. have designed a new antifouling peptide CPPPPNQNQNQNQDHWRGWVA (Figure 9) [38], including an anchoring domain (CPPPP), an antifouling domain (NQNQNQNQD) and a human immunoglobulin G (IgG) recognition domain (HWRGWVA). PANI nanowire was used as the supporter for the immobilization of the peptide by the specific covalently coupling reaction. The multifunctional peptide and PANI nanowire facilitated the specific identification of targets from serum. The detection limit for IgG in pure serum and real clinical samples is 0.26 ng/mL. Based on the self-assembled peptide and modified aptamer, Wang et al. developed an electrochemical detection method with high antifouling interface for detection of immunoglobulin in serum [39]. Due to the large specific surface area of porous gold and the high selectivity of aptamers, this electrochemical biosensor has high sensitivity and selectivity for immunoglobulin E (IgE). The linear range is 0.1 ~ 10 pg/mL, and the detection limit is low down to 42 fg/mL. Song et al. developed a sensitive electrochemical platform for the detection of T4 PNK [40], in which both the zwitterionic peptide CGGGGDKDKDKDK and sacrificial iron metal-organic framework (Fe-MOF) exhibited good antifouling performance by the synergistic effect. The biosensor had a wide linear range from 0.001 to 10 U/mL and a detection limit of 0.00035 U/mL.

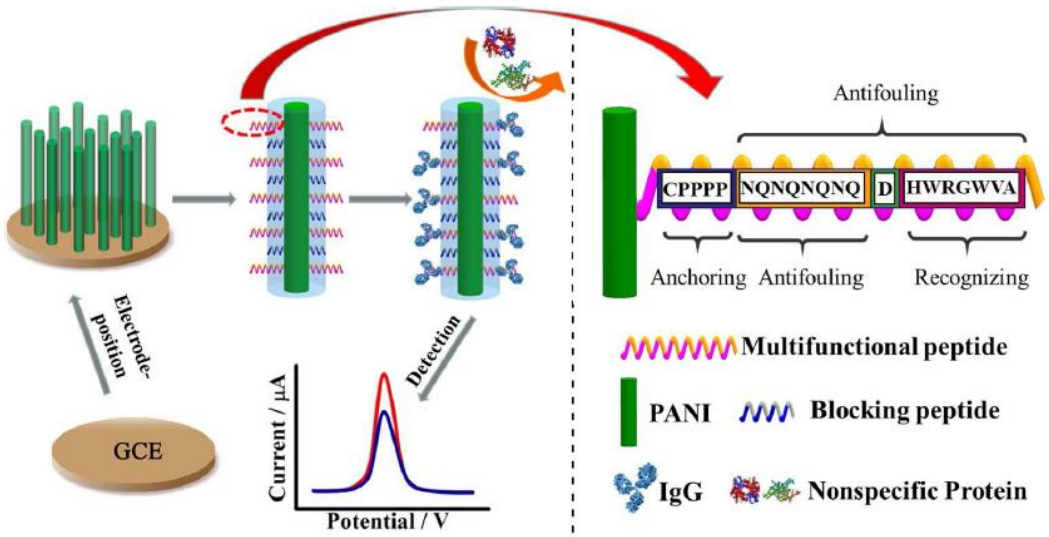

Figure 9. Schematic illustration of the preparation of a PANI supported peptides-based amperometric biosensor. Reprinted with permission from reference [38]. Copyright 2018 American Chemical Society.

2.4 PEGylated conductive films

Polyethylene glycol (PEG) is a non-toxic, high hydrophilic biocompatible polymer, which has been widely used to reduce the non-specific adsorption from proteins. It is considered as the "gold standard" of antifouling polymers for various biosensors. The antifouling properties of PEG are mainly affected by the surface packing density and polymer chain length. There are two methods to modify PEG: one is to adsorb the synthesized PEG on the electrode, and the other is to produce PEG *in situ* on the electrode. Both of these methods can endow the electrode with antifouling ability. However, PEG

directly modified on the electrode surface will affect the conductivity and structure of electrode to a certain extent. For this view, the grafting of PEG on conducting polymers such as PANI and Ppy has been reported for sensitive and antifouling interface in bioassays. For example, Zhang et al. designed a sensitive antifouling electrochemical biosensor for the quantitative detection of ATP with catechol&zwitterion-bifunctional β-PEG [41]. With the addition of zwitterions, β-PEG can effectively reduce the non-specific adsorption and improve the selectivity and sensitivity. The detection limit of the method is 0.01 pM, and the linear range is 0.01 pM ~ 10 nM. Wang et al. designed a sensitive electrochemical biosensor with an excellent antifouling performance based on PEG-polypyrrole (PEG-PPy) nanowires [42]. Jolly et al. used the PEG-PPy platform to enhance the antifouling ability of electrochemical biosensors (Figure 10) [43]. PPy was modified on the surface of gold electrode and PEG was mediated by electro-oxidation of amine group at one end of the PEG chain. Polyhistidine-modified aptamer was then connected to PEG-PPy by the Nα,Nα-Bis(carboxymethyl)-L-lysine ANTA/Cu^{2+} complex. The aptasensor was used to detect α-methylacyl-CoA racemase with a detection limit of 0.15 fM in buffer and 1.4 fM in human serum. Hui et al. prepared the PEG aniline (PANI/PEG) nanofiber to reduce the adsorption of non-specific protein and avoid the electrode pollution (Figure 11) [44]. The PANI/PEG electrode was used to determine the DNA biomarker of BRCA1 in serum with a detection limit of 0.0038 pM.

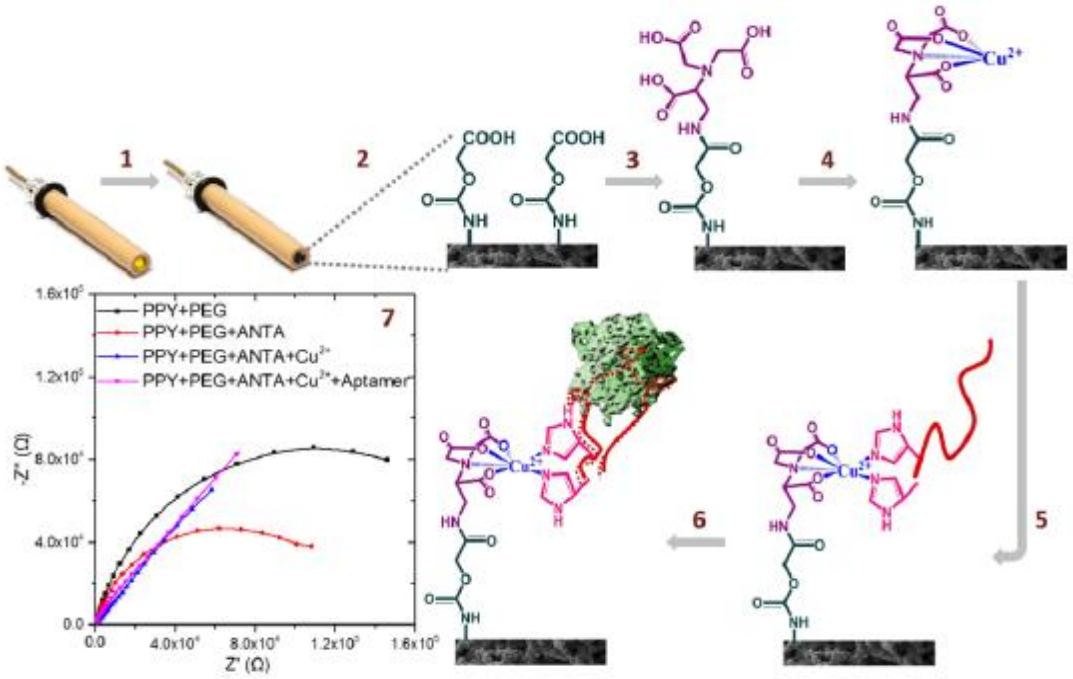

Figure 10. Schematic representation of aptasensor manufacture. Reprinted with permission from reference [43]. Copyright 2015 American Chemical Society.

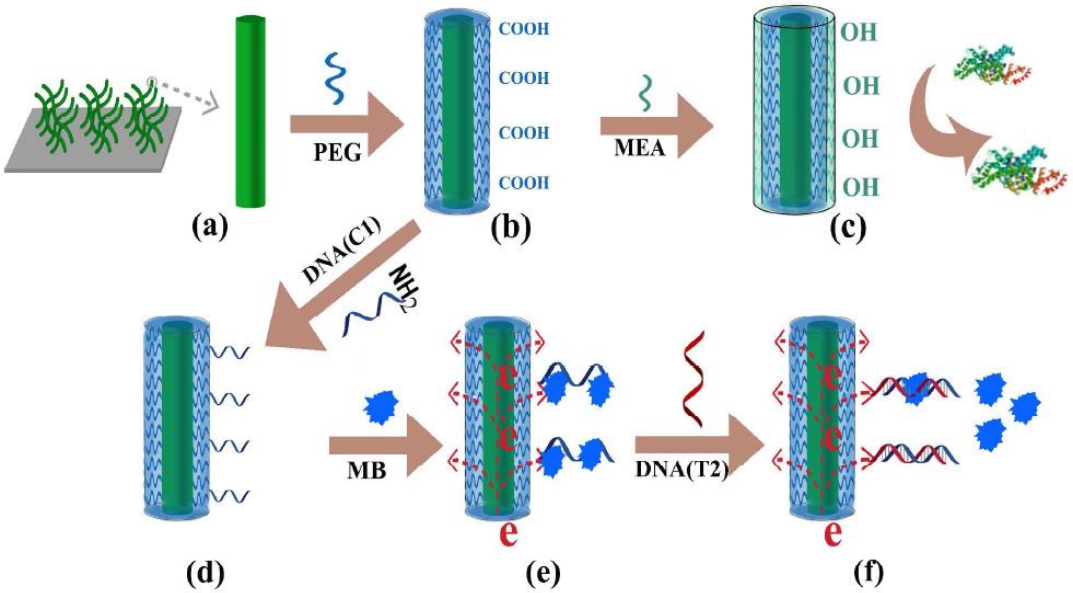

Figure 11. Illustration of the construction of PANI/PEG nanofibers antifouling interface and its application in ultrasensitive and low fouling DNA sensors. Reprinted with permission from reference [44]. Copyright 2015 American Chemical Society.

3. CONCLUSION

Great efforts have been made in the development of antifouling electrochemical biosensors. However, it is still desired to obtain a robust surface that can resist protein adsorption in complex biological media for clinical applications. This paper reviewed the progress in designing of effective antifouling SAMs for electrochemical biosensors. It should be valuable for the development of novel sensing devices with antifouling materials.

ACKOWLEDGMENTS
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