# Preparation and anti-biological adhesion performance evaluation of ZNO@PSBMA super hydrophilic coating

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In this study, zinc oxide (ZnO) nanoparticles were modified by means of dispersion using a γ-aminopropyl triethoxysilane coupling agent (KH550), obtaining KH550-ZnO. Then a certain amount of SBMA powder was put into the KH550-ZnO solution, and the ensuing polymerization reaction produced super hydrophilic PSBMA-KH550-ZnO powder. Finally, several polished aluminum sheets were immersed in the PSBMA-KH550-ZnO aqueous solution to start deposition. Upon completion of the deposition, the aluminum sheets were taken out and dried to obtain samples of the ZnO@PSBMA super hydrophilic coating. The structure, morphology and chemical composition of the powders and coatings were investigated by SEM, IR and EDS. The dispersion of KH550-ZnO in aqueous solution was analyzed. The results show that the particle size of KH550-ZnO decreases greatly in aqueous solution. The substrate can be superhydrophilic when deposited in PSBMA-KH550-ZnO aqueous solution for 130 minutes，After 48 hours of coating deposition, the compactness, roughness and friction resistance of the coating are greatly improved. ZnO@PSBMA superhydrophilic coating has good anti-protein, anti-bacterial and anti-algal adhesion properties.

(Received December 7, 2022; Accepted March 2, 2023)

*Keywords:* Anti-biological adhesion, PSBMA, Nano-ZnO

## 1. Introduction

Biological contamination can cause many hazards. Such as biological fouling will reduce the sailing speed of ships, increase fuel consumption, maintenance costs and release harmful gases [1-3]. At present, antifouling coating is one of the effective methods to solve biological fouling. After the prohibition of the use of organotin based coatings with lasting harm to the ocean in 2008, cuprous oxide and zinc-based self-polishing coatings with less toxicity occupy the largest market share, and the common organic antifouling coatings such as copper pyridyl thione (CuPT), zinc pyridyl thione (ZnPT) and heterocyclic fungicides are mainly [4]. However, studies have found that the extensive

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https://doi.org/10.15251/DJNB.2023.181.315

use of these inorganic tin self-polishing antifouling agents will also lead to the accumulation of toxic and harmful substances in the Marine environment and harm the ecological environment [5-9]. In order to overcome the adverse effects of traditional Marine antifouling agents, all countries are actively looking for a new type of environmental protection and high performance antifouling agent.

Zwitterionic polymer refers to the polymer compound with the same positive groups (such as -NH4+) and negative groups (such as -PO4-,-SO5-) in the monomer molecule [10, 11]. They can interact with water molecules through strong electrostatic interaction and hydrogen bonding to form a dense hydration layer bonded with water. Therefore, amphoteric ion molecules are considered as ideal antifouling materials [12, 13]. Sulfobetaine methacrylate (SBMA) is a small molecular monomer with -NH4+ and -SO5- side chains, and its polymerization will generate macromolecule polysulfobetaine (PSBMA). Many studies have shown that PSBMA has excellent antifouling function in inhibiting protein and bacterial adhesion in aqueous media [14-19].

In addition, nano-ZnO is a semiconductor with significant application in different fields, which has the characteristics of non-toxic, chemically stable, low price and wide band gap [20]. Because of its photocatalysis, it has microbial activity and degrades many toxic pollutants [21, 22]. Even in the absence of light, Zn+ can be released in the aqueous solution of biofilms such as bacteria, which can be inactivated by the REDOX reaction between Zn+ and the membrane surface of bacteria [23, 24]. Therefore, a simple ZnO hydrophilic antifouling coating can be designed to achieve antifouling effect by combining the dual efficacy of nano-ZnO sterilization and anti-adhesion of amphoteric polymer hydration layer.

In this study, zinc oxide (ZnO) nanoparticles were modified by means of dispersion using a γ-aminopropyl triethoxysilane coupling agent (KH550), obtaining KH550-ZnO. Then a certain amount of SBMA powder was put into the KH550-ZnO solution, and the ensuing polymerization reaction produced super hydrophilic PSBMA-KH550-ZnO powder. Finally, several polished aluminum sheets were immersed in the PSBMA-KH550-ZnO aqueous solution to start deposition. Upon completion of the deposition, the aluminum sheets were taken out and dried to obtain samples of the ZnO@PSBMA super hydrophilic coating. The coating samples prepared in this way were put under various tests to study the dispersion of KH550-ZnO in aqueous solution, the species and composition of the functional groups on the surface of the coating, the impact of deposition time on wettability, and the performance of the coating in resisting protein, bacteria, and algae growth.

## 2. Experimental

### 2.1. Materials

Nano zinc oxides (nano-ZnO,30nm) was purchased from Guangzhou Changyu Chemical Technology. γ-aminopropyltriethoxysilane (KH550) was purchased from Nanjing Chuangshi Chemical additives. Sulobetaine methacrylate (SBMA), sodium bisulfite and potassium persulfate were purchased from Aladdin Shanghai. Ethyl alcohol was purchased from Sinopharm Chemical Reagent Co., LTD. Escherichia coli (CMCC44102) was purchased from Shanghai Bioresource collection center. LB Nutrient Agar was purchased from Qingdao Hope Bio-Technology. Phosphate Buffered Saline (PBS) was purchased from Guangzhou Hewei Pharmaceutical Technology Co., LTD. Fluorescein isothiocynate-labeled bovine serum albumin (BSA-FITC) was purchased from Beijing Bersee Science and Technology Co., LTD. 1060 pure aluminum plate was purchased from Yonghao

Hardware insulation material store. Marine green alga was purchased from Green Water transfer store.

### 2.2. Modification of nano-ZnO

Mix 3.0 g nano-ZnO powder, 50.0 mL ethanol and 5.0 mL deionized water, stir the solution and apply an ultrasonic treatment. Add 225 KH550 to the above solution with a pipette gun and stir the solution using a heat-collecting constant-temperature water bath (600 rpm, 40℃) for 4 hours. Filter the modified mixture with a suction filter, wash the resulting mixture thrice with ethanol (removing most impurities), dry it with blast air oven, and grind it to obtain KH550-ZnO powder.

### 2.3. Preparation of ZnO@PSBMA coating

Firstly，mix 3.0 g SBMA, 3.0 g KH550-ZnO and 30.0 mL deionized water, stir the solution, and apply an ultrasonic treatment. secondly, add 3.0 g potassium persulfate and 0.06 g sodium bisulfite to the solution, stir the solution using a heat-collecting constant-temperature water bath (600 rpm, 65℃)for 8 hours. Thirdly, put the stirred solution into the dialysis bag and perform dialysis in deionized water to remove impurities (48 hours, replacing the pure water every 12 hours). Fourthly, perform freeze drying on the dialyzed solution for 72 hours to obtain super hydrophilic PSBMA-KH550-ZnO powder, the procedure of preparing PSBMA-KH550-ZnO powder is shown in Fig 1. Finally, mix the PSBMA-KH550-ZnO powder with deionized water, stir the solution, and put the polished 1060 pure aluminum sheets into the solution to deposition. After a certain number of hours, the samples of ZnO@PSBMA super hydrophilic coating are obtained.

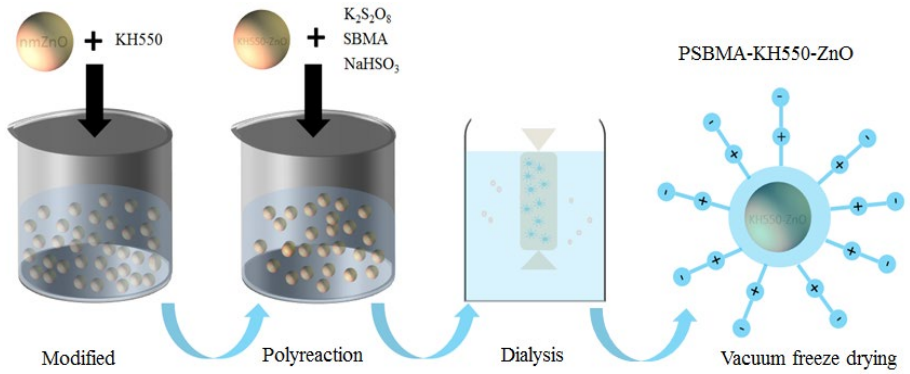


*Fig. 1. Procedure for preparing PSBMA-KH550-ZnO powder.*

### 2.4. Material characterization

The contact angle of the coating is tested using the contact angle measuring device (DSA30, KRüSS, Germany). The morphology of the coating is characterized using a high-resolution field emission scanning electron microscope (Sigma 500, Carl Zeiss, Germany). The functional groups of the samples are characterized using Fourier transform infrared spectrometer (Nicolet IS10, Thermo Fisher Scientific, America), measurement range: 750~4000 cm-1. The elemental composition and content of each element of the outer layers of powder and coating are characterized using X-ray spectroscopy. The dispersed particle size of the sample in solution was characterized by laser particle size analyzer (NKT6100-B, NKT, China).

### 2.5. Cross hatch tape adhesion test and abrasion test

The abrasion of coating is tested on a steel velvet abrasion machine. Mount the abrasion rod of the steel velvet abrasion machine to the back side of the sample of coating, attach 600-mesh sandpaper to the coated side of the aluminum sheet, and glue the other side of the sandpaper to the table. Apply a 100 g weight and start the abrasion test (2 m/min). Measure the wettability of the coating each time the abrasion rod slides 1 m.

### 2.6. Anti-biological testing of coating

Fluorescein isothiocynate-labeled bovine serum albumin (BSA-FITC) was diluted in phosphate buffer solution (PBS), and the concentration of the resulting solution was 1 mg mL−1. The samples were then incubated in BSA-FITC solution in a dark room for 24 h at 37 °C followed by thorough rinsing with PBS solution. The BSA proteins adsorbed on samples was detected using a fluorescence microscope (Eclipse, Nikon, Japan). The fluorescence intensity weaker, the anti-protein adhesion ability stronger.

Escherichia coli (CMCC44102) is used as the bacterium strain for the antibacterial testing; sanitary polyethylene (HDPE) is used as the blank control sample to evaluate the antibacterial rate of the coating sample. Wipe the surfaces of sample with alcohol for sterilization, and wash them with deionized water. In a sterile environment, fetch a certain amount of third-generation inclined bacteria with a sterile inoculation ring, add the bacteria into 30.0 mL of PBS buffer and shake the liquid. Put the sample into a sterilized petri dish, fetch 100 μ L of the bacterial solution with a pipette gun, and drop the solution onto sample, Cover the same sample on the surface of the sample so that the strain is in full contact with the surface. Incubate the sterilized petri dish in an incubator (24 h, 37℃). After incubation, Immerse the samples in a sampling cup filled with 50.0 mL of PBS buffer, elute the bacteria using a shaker (10 min), and collect the bacteria in the PBS buffer. Take 200 μ L of the bacterial solution dilute in 1.8 mL of PBS buffer to obtain a diluent 10-1 bacterial solution. Repeat this operation 6 times to obtain diluent bacterial solutions from 10-1 to 10-6. Then take bacterial solutions drop onto the L B Nutrient Agar, and apply the liquid evenly with a coating rod. Place the L B Nutrient Agar down in a constant-temperature incubator (37℃) until clear colonies appear. Three parallel samples were made for each sample, and the colony number was averaged. The antibacterial rate is calculated using the formula of $R = (B - A)/B \times 100\%$, where R (%) is the antibacterial rate, A is the number of colonies on the coating, and B is the number of colonies on the surface of the control sample.

Marine green algae is used as the algae species in the anti-algae adhesion testing. Add 50 mL of marine green algae into the container and cultivate it at room temperature. Immerse the coating and control samples in the algae-containing water. After a period of time, take out the sample to observe the surface.

## 3. Results and discussion

### 3.1. Analysis of particle size, FTIR characterization and preparation mechanism

As with any ultrafine powder, nano-ZnO has a high specific surface area and surface energy and is prone to agglomeration. This makes it difficult for the particles to perform their functions as expected in practical use, so it is necessary to weaken the agglomeration tendency by modifying the

surface of the particles. KH550 is a good surface modifier for inorganic particles and can be used for surface organic modification of nanoparticles. In this experiment, KH550 was grafted onto nano-ZnO to improve the agglomeration phenomenon of nano-ZnO.

Fig. 2 shows the particle size distribution of the sample in solution. As can be seen, the dispersed particle size of nano-KH550-ZnO compared with nano-ZnO in solution is greatly improved. This is largely due to the KH550 molecules are first hydrolyzed to form Si-OH bonds in the ethanol-water mixture,, and then form the Si-O-Zn chemical bond with the -OH on nano-ZnO through dehydration and condensation, which weakens the agglomeration tendency of the particles. Figure 3(a) shows the process of grafting KH550 onto nano-ZnO.

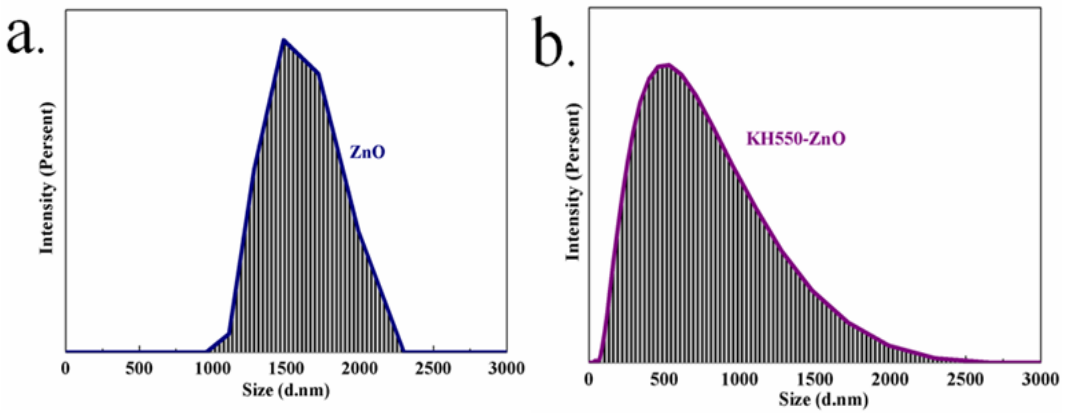


*Fig. 2. (a) Particle size distribution of nano-ZnO. (b) Particle size distribution of nano-KH550-ZnO.*

After the modification of nano-ZnO is completed, the monomer SBMA molecules undergo a polymerization reaction to produce PSBMA-KH550-ZnO nanoparticles in the nano-KH550-ZnO solution. Fig 3(b) and 3(c) shows the process of Polymerization reaction. Because the side chain of each monomer of SBMA contains a positive group quaternary amine (-NC4+) and a negative group sulfonate (-SO3-), the surface of PSBMA-KH550-ZnO nanoparticles has a large amount of 1:1 positive and negative charges. PSBMA-KH550-ZnO nanoparticles can form a hydration layer in water because of the strong interaction between water molecules and the positive and negative groups of the side chain. In this case, PSBMA-KH550-ZnO nanoparticles surface will form a superhydrophilic physical barrier to prevent biological adhesion.

Fig 3(d) shows the FTIR characterization of nano-zno and PSBMA-KH550-ZnO nanoparticles samples. As shown in the figure, there are many new peaks in nano-PSBMA-KH550-ZnO compared with nano-ZnO. 1,722 cm-1 is the stretching vibration peak of ester carbonyl in PSBMA. 1,480 cm-1 corresponds to the quaternary amine (NC4+) on PSBMA. 1,166 cm-1 is the stretching vibration peak of C-O-C on PSBMA. 1,036 cm-1 is the stretching vibration peak of S=O on PSBMA. 929 cm-1 and 793 cm-1 are the peaks of the bonds of Si-O-Zn and Zn-O, respectively. These new characteristic peaks indicate the successful preparation of PSBMA-KH550-Zno nanoparticles.

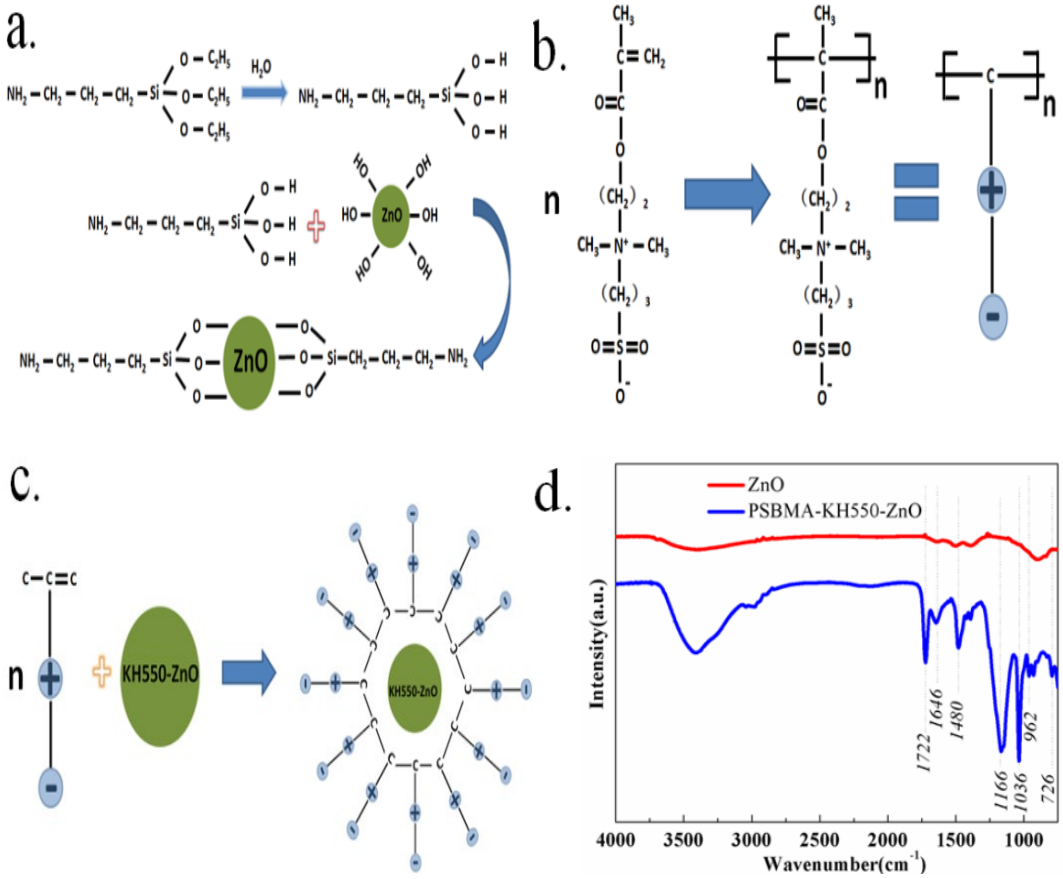


*Fig. 3. (a) The process of grafting KH550 onto nano-ZnO. (b) SBMA polymerization process. (c) PSBMA-KH550-Zno nanoparticles preparation process. (d) FTIR characterization of nano-ZnO and PSBMA-KH550-ZnO nanoparticles samples.*

### 3.2. Impact of deposition time on coating properties

The impact of deposition time on the wettability of the coating surface is shown in Fig 4(a).It can be seen from the figure that after 120 minutes of deposition on the polished aluminum substrate surface, the coating becomes completely hydrophilic.

The impact of deposition time on the abrasion of coating surface is shown in Fig 4(b). Under the same abrasion conditions, it can be seen that the longer the coating deposition time, the smaller the increase of contact Angle. After abrasion of 10 m, the contact Angle of the coating after 48 hours of deposition rises to about 20° .

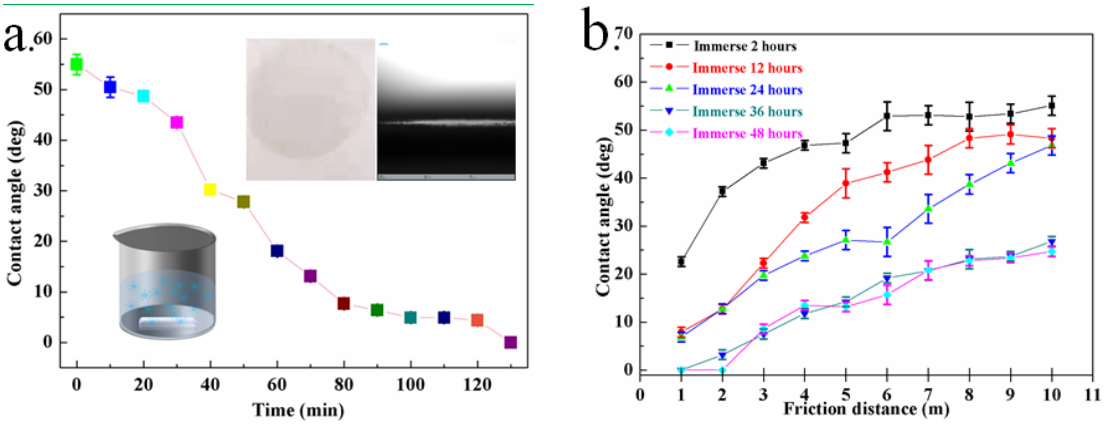


*Fig. 4. (a) The impact of deposition time on the wettability of the coating surface. (b) The impact of deposition time on the abrasion of coating surface.*

### 3.3. Analysis of micro morphology of coating

Fig 5 shows SEM images of the surface morphology of the ZnO@PSBMA coating. Fig 6(a1), (a2) and (a3) shows the surface SEM of the coating deposited for 24 h, it can be seen that there are many nanoparticles deposited on the surface, and the rough structure is obvious, but the surface composition and structure are loose. Fig 6(b1), (b2) and (b3) shows the surface SEM of the coating deposited for 48 h, it can be seen that there are still many uniform and uneven rough structures on the surface of the coating, but compared with the coating deposited for 12 h, it looks more compact and stable between the surface composition and structure. This can show that no matter how long the coating is deposited, the surface has micro/nano rough structure, which is one of the necessary conditions for constructing super hydrophobic surface. But the deposition time is long, the surface structure is more compact and stable.

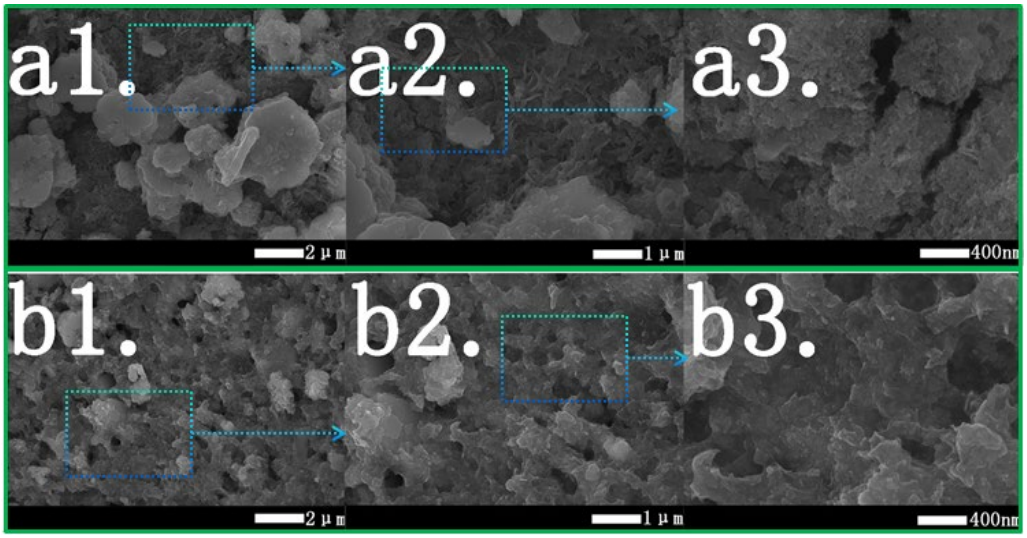


*Fig. 5. Micro-morphology of the surface of the PSBMA@ZnO coating.*

Figure 6 shows several element distribution images of the ZnO@PSBMA coating obtained through EDS surface scanning. The elements Zn, C, S, O and N are important constituent elements of the coating. It can be seen from the element distribution images that these five elements are evenly distributed, which indirectly proves that the coating surface components are evenly distributed together.

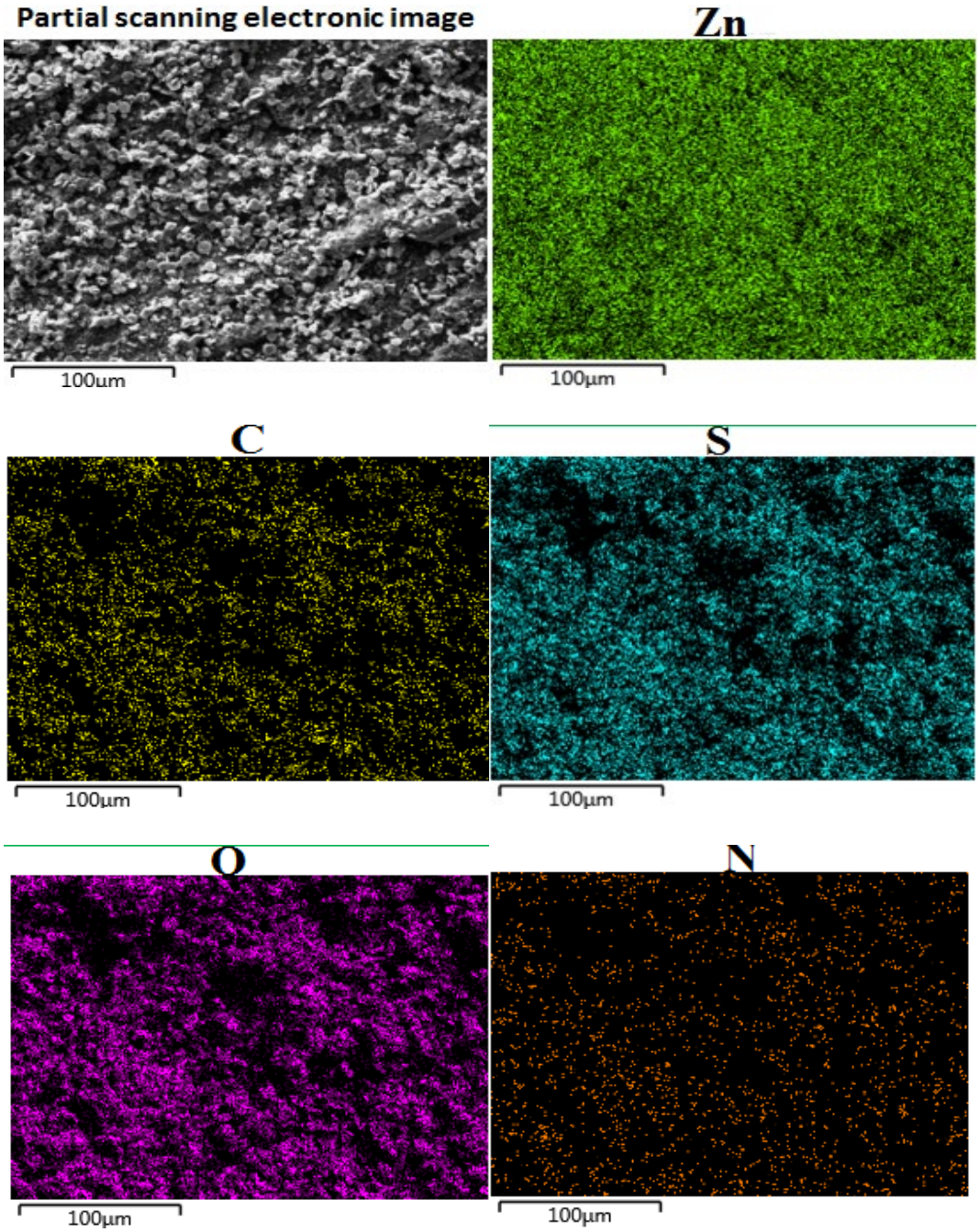


*Fig. 6. EDS local surface scanning spectrum of ZnO@PSBMA coating.*

### 3.4. Analysis of the biology resistance of the coating

Protein adhesion is one of the important factors in the formation of biological adhesion on material surface, which can be determined by surface wettability. In this work, Fluorescein isothiocynate-labeled bovine serum albumin (BSA-FITC) was selected as a model protein to investigate the protein adsorption. Fluorescence image of sample surface after incubation in BSA-FITC solution is shown in Fig 7(a). As can be seen from the figure, the fluorescence signal of ZnO@PSBMA coating deposited for 24 h and 48 h is much weaker than that of pure Al surface，it can indicate that ZnO@PSBMA coating surface has good anti-protein adhesion property. This can be explained by the characteristics of the coating material. On the surface of the particles formed after the nano-ZnO molecules are wrapped by Zwitterionic polymers, there are a large number of side chains in which the ratio of positive charges to negative charges is 1:1. As a result, the side chains of the positive and negative groups interact strongly with water molecules, thus forming a protective layer of water film that makes it difficult for protein molecules to adhere to the material surface.

Bacteria are an important part of the biofouling that occurs on material surface. In the experiment of this study, the common gram-negative Escherichia coli were used to test the antibacterial performance of the coating. A sanitary-grade polyethylene (HDPE) board was used as the blank control to compare with the polished aluminum sheet. The results of the antibacterial test are shown in Table 1. As can be seen from Fig. 7（b）, a large number of colonies of the listed three bacteria strains grew on the culture medium of the blank control and the polished aluminum sheet. In contrast, almost no colonies can be found on the culture medium of the ZnO@PSBMA coating samples. According to the antibacterial rate formula $R= (B - A)/B \times 100\%$. The antibacterial rate of the polished aluminum sheet is only 24.4%. The antibacterial rate of the ZnO@PSBMA coating reaches 93.3%. This contrast demonstrates that the ZnO@PSBMA coating is excellent at resisting Escherichia coli. This is mainly due to the water film protective layer of superhydrophilic surface and the antibiological activity of nano-ZnO.

*Table 1. The results of the antibacterial test.*

| Sample | Colony count | Antibacterial rate |
|---|---|---|
| ZnO@PSBMA coating | 6 | 93.3% |
| Aluminum | 68 | 24.4% |
| HDPE | 90 | - |

Algae adhesion is the most common form of biological adhesion. In the experiment of this study, marine green algae were used for the anti-algae adhesion testing. As shown in Fig 7(c), aluminum has adhered to a large number of green algae on its surface after 30 and 60 days in green algae, while ZnO@PSBMA coating has not adhered to green algae on its surface after soaking for 30 days. After soaking for 60 days, trace amounts of green algae adhered to the surface，it is due to the coating hydrophilic failure, the disappearance of the protective layer. These phenomena indicate that the coating can resist algal adhesion for a certain period of time. Over time, the coating will fail due to its superhydrophilicity, resulting in surface adhesion to chlorella. As time goes on, the coating will lose its superhydrophilic ability and the surface will adhere to green algae.

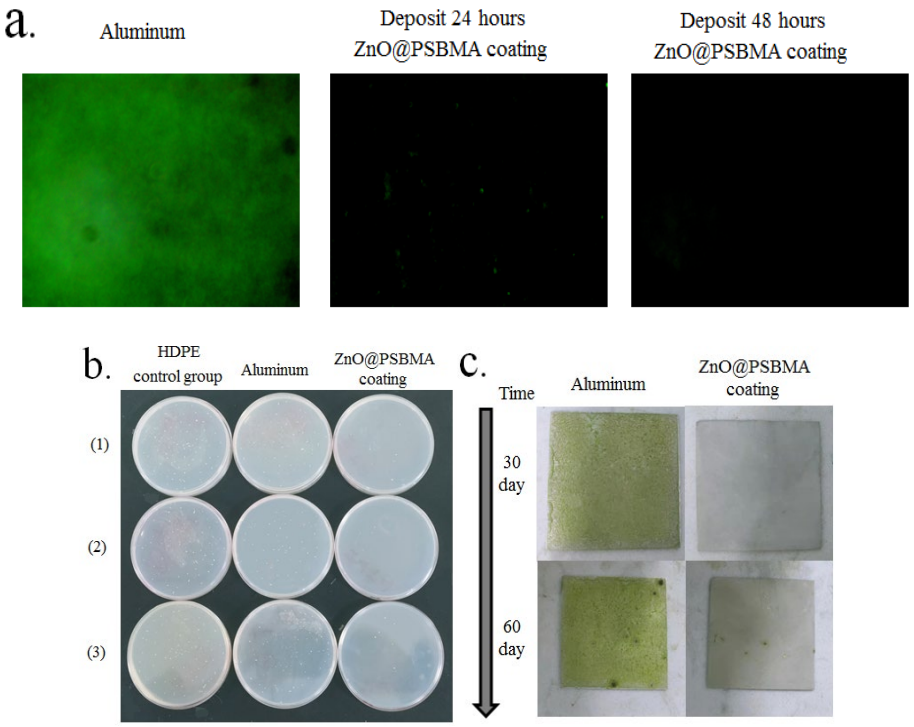


*Fig. 7. (a) Fluorescence image of sample surface. (b) The results of the antibacterial test. (c) The results of the anti-algae adhesion test.*

### 3.5. Mechanism analysis of anti-biological adhesion on coating surface

#### *3.5.1. Corrosion resistance test*

Fig 8 is a schematic diagram of anti-biological adhesion mechanism of nano-ZnO@PSBMA superhydrophilic coating. On the coating of surface covered with PSBMA-KH550-ZnO particles, there are a large number of side chains which the ratio of positive charge to negative charge is 1:1. As a result, the side chains on the positive and negative groups interact strongly with water molecules, thus forming a strong protective layer of water film on the particle surface and making it difficult for organisms to adhere to the coating surface. In addition, combined with the anti-biological activity of nano-ZnO, a double anti-biological adhesion effect can be produced to achieve the anti-biological adhesion effect.

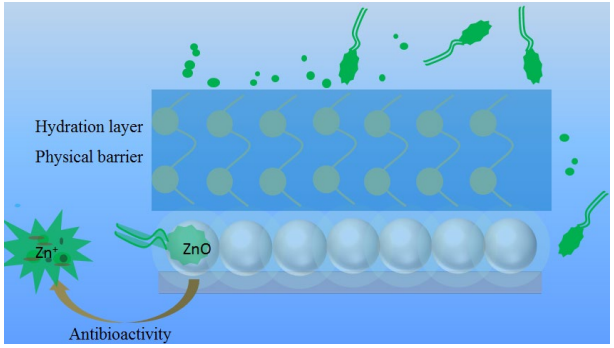


*Fig. 8. The schematic diagram of anti-biological adhesion mechanism of ZnO@PSBMA superhydrophilic coating.*

## 4. Conclusions

The purpose of this study is to reduce biological adhesion on material surface by combining the anti-biological activity of nano-ZnO with the anti-biological adhesion properties of hydrophilic surface, providing a basic research and a new idea for preparation of anti-biological adhesion coating. In this study, KH550-ZnO was prepared by modifying nano-ZnO with KH550. As a result, the particle size of KH550-ZnO in solution was greatly reduced. Superhydrophilic PSBMA-KH550-ZnO powder was prepared through a polymerization reaction by SBMA with nano-KH550-ZnO. Finally, the ZnO@PSBMA coating was successfully prepared on a polished aluminum substrate through deposition. The coating can attain superhydrophilicity after 130 minutes of deposition. The coating surface stability, abrasion resistance and adhesion are the best after 48 hours of deposition. The most important is the ZnO@PSBMA coating has a strong ability to resist the adhesion of proteins, bacteria and algae by test. Overall, our current work reported a new strategy for reducing biological adhesion on material surface, implying important application in the field of environmentally friendly coatings.

**Acknowledgements**

This work was supported by National Natural Science Foundation of China (52073127) and Changzhou Sci & Tech Program (CM20193004).

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