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Zwitterionic SiO_2 nanoparticles exhibit the great potential for improving the antifouling performance of hydrophobic PVDF membrane.

Journal Name RSCPublishing

ARTICLE

Zwitterionic SiO_2 nanoparticles as novel additive to improve the antifouling property of PVDF membrane

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Hybrid polyvinylidene fluoride (PVDF) ultrafiltration (UF) membranes with excellent antifouling property were prepared by non-solvent induced phase separation though blending zwitterionic SiO_2 nanoparticles. Lysine was used to modify SiO_2 nanoparticles to generate surface zwitterion with the amino acid type. Zwitterionic SiO_2 nanoparticles could be distributed uniformly in the membrane bulk to avoid massive agglomeration, and significantly improved the hydrophilicity and separation performance of PVDF UF membrane. The amount of BSA adsorbed on hybrid ZP-5% membrane surface of static fouling test decreased to $10 ug/cm^2$ and the secondary water flux recovery rate (FRR) increased to more than 95% for the dynamic antifouling test of BSA and HA, the addition of zwitterionic SiO_2 nanoparticles could enhance the antifouling ability though inhibiting the irreversible fouling to prolong the service life of PVDF UF membrane.

1. Introduction

In recent years, membrane technology has attracted a lot of attention and plays an important role in water treatment, industrial, pharmacy, food and other fields.[1] However, the membrane fouling problem is inevitable for the practical application of all the membrane separation processes, leading to the increase of application costs and reduction of service life. Therefore, the preparations of low fouling or non-fouling membranes have been the hot issues in the field of membrane modification at this stage.[2-5]

Numerous studies have demonstrated that hydrophilic modification of hydrophobic membrane is an effective way to improve the fouling resistance properties. As we all know, water molecules are preferentially adsorbed at the hydrophilic interface during separation process to form a hydration shell, which can effectively block the hydrophobic pollutants to contact with the membrane surface and alleviate subsequent deposition of pollutants, enhancing the antifouling performance of the hydrophobic membrane.[6-10] Zwitterionic materials with biomimetic properties is considered as one of the best fouling resistant materials due to the excellent hydration capacity based on the positive and negative groups with strong hydrophilicity, water molecular can be priority adsorbed to form protective layer in the application process.[11-15] However, the strong hydrophilicity of zwitterionic materials limit their solubility in conventional organic solvents, so zwitterionic materials

can't be directly blended with commercially available hydrophobic membrane materials. Therefore, the convenient use of zwitterionic materials remain a challenge, the selected method of surface grafting and preparation of block copolymers with harsh conditions and multi-step reaction still limit the large-scale industrial applications of zwitterionic materials.[16-19]

Hydrophilic SiO_2 nanoparticles with the large surface area and strong hydrophilic features are usually used as modified additives for coatings, pharmaceuticals, environmental protection, separation membranes and other fields.[20-23] In addition, due to the particle surface covered with reactive active sites, SiO_2 nanoparticles are ideal carrier of functional materials, but agglomeration is the major factor of limiting the application of nanomaterials.[24-28] Zwitterionic nanoparticles can be prepared though surface modification, which will overcome the demerits for application such as agglomeration problem of nanomaterials and insolubilization of zwitterionic materials in organic solvent. Using SiO_2 nanoparticles with good dispersibility as carrier, makes it possible to enhance fouling resistance of the hydrophobic materials with zwitterionic medium.

Recently, studies have found that amino acids are potentially zwitterionic materials, which have a wide range of sources, excellent design characteristics and top antifouling effect.[29-34] Therefore, in this paper lysine was used for modifying the SiO_2 nanoparticles to generate zwitterionic surface and hybrid PVDF UF membranes were prepared by blending this new additive of zwitterionic nanoparticles (ZP), and the effect of adding zwitterionic nanoparticles for the

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separation performance and fouling resistant properties were investigated of PVDF membrane. This paper aims to provide a convenient method for the preparation and large-scale production of antifouling PVDF membranes using zwitterionic materials.

2. Experimental

2.1 Materials

PVDF (MG105) was purchased from Arkema, Bovine serum albumin (BSA, 67000Da), PEG (Mw~20000) and lysine were purchased from Sinopharm Chemical Reagent Co. (China). Humic acid (HA, Fulvic acid > 90%), 3-glycidyloxypropyl (dimethoxy) methylsilane (GPMS, KH560), and hydrophilic SiO_2 nanoparticles (30nm) were purchased from Aladdin Co. (China), ethanol, hydrochloric acid and dimethylacetamide (DMAC) were purchased from the local corporation with commercially analytical grade. All the reagents were used without any purification.

2.2 Modification of SiO_2 nanoparticles

1g SiO_2 nanoparticles are dispersed into 20% (v/v) alcohol aqueous solution by ultrasound for 30 min, then 0.3g GPMS and 1 drop of hydrochloric acid were added into the above solution to react for 6 hours under magnetic stirring at 60℃. After that, 5ml lysine aqueous solution (10%) was added into the SiO_2 dispersion solution and kept for another 3h to react, finally, the reaction products were centrifuged, washed and dried for subsequent characterization and modification.

2.3 Membrane preparation

A certain amount of modified SiO_2 nanoparticles was added to the mixed solution of DMAC 40g and H_2O 10g to ultrasound for 1h to promote the dispersion of nanoparticles, 5g PEG was added to further induce the uniform dispersion of modified SiO_2 nanoparticles under magnetic stirring for 2h. 10g PVDF powder was added to the above solution and dissolved completely as membrane casting solution to prepare PVDF hybrid membranes by non-solvent induced phase separation (NIPS) method with ice water as coagulating bath, the formed PVDF membrane were placed in water more than 12h to remove excess solvent and porogen. The dosages of modified SiO_2 nanoparticles were 2% and 5% for the PVDF mass and prepared hybrid membranes were labeled ZP-2% and ZP-5%, respectively. The PVDF membrane with 5% addition of pristine SiO_2 was used as reference sample.

2.4 Characterization

Particle size distribution and zeta potential of SiO_2 nanoparticles were tested by nanoparticle size and potential analyzer (Nano ZS, Malvern). The chemical compositions of modified SiO_2 nanoparticles were characterized by fourier transform infrared spectroscopy (FTIR, Nicolet 8700) and X-ray photoelectron spectroscopy (XPS, Shimadzu AXIS UltraDLD). Attenuated total reflectance fourier transform infrared spectroscopy (ATR-FTIR, Nicolet 8700, USA) was used to examined the surface composition of PVDF membranes, the membranes were analyzed on a ZnSe crystal as the internal refection element with an aperture angle of 45°, the ATR-FTIR spectra were recorded by the accumulated average of 32 scans at 4cm^{-1} resolution. Surface hydrophilicity of neat and modified membranes was evaluated by water contact angle (CA, OCA40Micro, Germany) on the membranes surface at room temperature. A total of about 3 μl of deionized water was dropped onto the membrane surfaces using a micro syringe and the results were obtained using the drop shape image analysis system to evaluate the surface wetting ability. In addition, the wetting property of membrane matrix (interior of the membrane) was examined though a novel method. Hybrid membrane without skin layer was directly obtained using casting solution by spin coating (5000 rpm/min)and thermosetting (80℃), which imitated the membrane matrix structure. Then contact angle can be directly used to test the hydrophilicity of mimetic matrix with the same method of membrane surface. Surface and cross-section morphology of membranes were viewed with the field emitting scanning electron microscope (SEM, Hitachi SU8010, Japan), all the samples were coated with gold before observation and cross-section of membranes were acquired using liquid nitrogen. The surface microstructure of the membranes was characterized with Agilent5500 atomic force microscope (AFM, Agilent Technologies Inc., USA) in the tapping mode, and the roughness of membranes were characterization with Rq value.

2.5 Filtration

The separation performance of pristine and hybrid PVDF membranes were investigated using a dead-end filtration system with affective membrane area of 12.5 cm^2. The filtration experiments were carried out with the compacted pressure at 0.15MPa for 30 min using pure water as feed solution, and the pressure was lowered to 0.1MPa to obtain a stable flux by comparing the flux values every 5 minutes. The pure water flux was noted as J_1 (L/m^{2}h) and calculated by the following equation (1).

The rejection of all membrane was tested with BSA solution (1g/L, pH7.4) as feed solution. The experiment process was referred to the water flux test method and the permeate concentration was examined via UV spectrophotometer (UV-1800, Shimadzu) with the characteristic wavelength at 280 nm, the rejection was calculated as the following equation (2).

$$\mathrm{J} = V/(A \times t) \qquad (1)$$

$$\mathrm{R} = (1 - C_P/C) \times 100\% \qquad (2)$$

Where V (L) is the volume of permeated water, t (h) is the permeation time and A (m^2) is effective area for filtration, Cp and C are contributed to the BSA concentrations of permeate and feed solution.

2.6 Fouling test

The static fouling test was carried out by immersing the hybird PVDF membranes (3×3 cm^2) in pollutant solution (BSA, 0.5g/L, pH7.4) without additional pressure-assisted BSA into the membrane pore. After adsorption-desorption equilibrium for 12h at room temperature, the concentrations of BSA solution before and after the adsorption of PVDF membranes were measured with UV-1800 and the mass (μg/cm^2) of adsorbed BSA on the membrane surface was calculated. In order to test the stability of nanoparticles during separation process in the aquatic environment, the water flux of long time (8h) of hybrid membrane was tested.

The dynamic fouling experiments were executed using a circulating mode with alternate feed solution of pure water and pollutants solution. BSA and HA are used as pollutants feed solution, respectively. Firstly, stable pure water flux was recorded as J_1, then the feed was replaced with pollutants solution and obtained stable flux was recorded as J_p, after that, the tested membranes were rinsed with pure water for 20min and secondary pure water flux was obtained as J_2.Water flux recover ratio (FRR) values and irreversible fouling resistance (IFR) values were calculated by the followed equation (3) (4). In addition, the dynamic fouling experiment was repeated several times to evaluate the antifouling stability of zwitterionic nanoparticles in membrane matrix.

$$\mathrm{FRR} = \mathrm{J}_2/\mathrm{J}_1 \times 100\% \qquad (3)$$

$$\mathrm{IFR} = 1 - \mathrm{FRR} \qquad (4)$$

3. Results and discussion

Zwitterionic modification method of SiO_2 nanoparticles was shown in Fig. 1, the silane coupling agent KH560 was used to react with SiO_2 nanoparticles under acidic condition to prepare the intermediate products with active epoxy groups, according to the ring-opening reaction between the amino group of lysine and epoxy groups on the SiO_2 particle surface, lysine with a pair of amino and carboxyl was grafted onto SiO_2 nanoparticles to obtain zwitterionic nanoparticles with amino acids type.

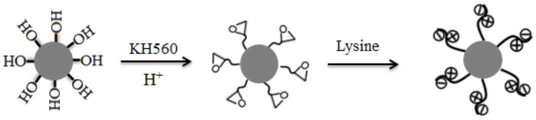

Fig. 1 Scheme for the zwitterionic modification of SiO_2 nanoparticles

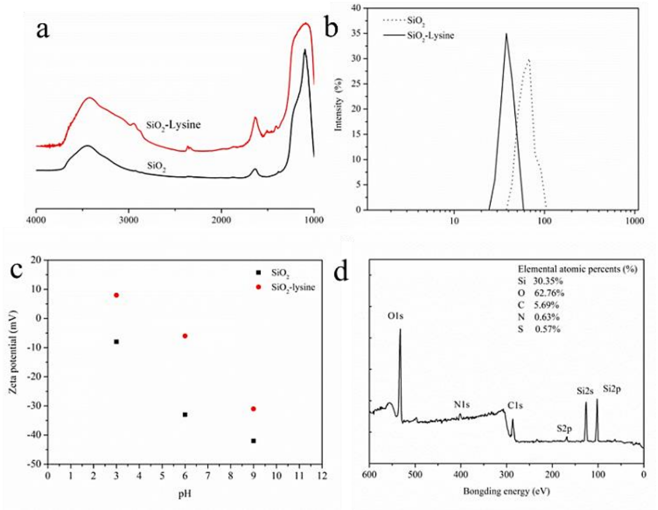

Fig. 2 Characterization of pristine and lysine modified SiO_2 nanoparticles (a: FTIR, b: Particle size, c: Zeta potential, d: XPS of SiO_2-lysine)

FTIR spectra of SiO_2 nanoparticles before and after modification were shown in Fig. 2a, comparing the spectra of pristine and lysine-modified SiO_2 nanoparticles, three new peaks at 1410 cm^{-1},1510cm^{-1} and 2960cm^{-1} were observed , which were contributed to the plane bending vibration absorption peak of OH bond belong to carboxyl, shear vibration absorption of amino and methylene absorption peak. Meanwhile hydroxyl absorption at 3400cm^{-1} peak was significantly broadened, suggesting the increase of carboxyl and amino groups on the surface of SiO_2 nanoparticles, All these results indicate that the lysine with amino group and carboxyl group were successfuly grafted onto the SiO_2 nanoparticle surface and novel modified SiO_2 nanoparticles were prepared with zwitterionic surface.

To investigate the effect of zwitterionic modification for the agglomeration problem of SiO_2 nanoparticles, the particle size distribution of SiO_2 nanoparticles before and after modification were shown in Fig. 2b. It was clearly observed that the size distribution of modified SiO_2 nanoparticles range was more narrowed and the average diameter become smaller than pristine SiO_2 nanoparticles, the average diameter decreased from 80nm to about 40nm, which was close to the diameter (30nm) of the pristine material SiO_2 nanoparticles, it could be known that the agglomeration problem of SiO_2 nanoparticles with zwitterionic modification was significantly alleviated. The appropriate reason should be attributed to surface modification strengthening the interface stability of nanoparticles, the ionogenic pair of amino and carboxyl groups reduce the absolute value of nanoparticles surface charge, leading to the better dispersion effect.

Because lysine included a pair of positive and negative ions, the zeta potential of modified SiO_2 nanoparticles was tested (Fig. 2c), obviously, the SiO_2-lysine exhibit negative charge at high pH and positive charge, which is a characteristic of zwitterionic materials. The characteristic elements of N and S belonging to lysine were detected in the XPS spectra of SiO_2-lysine nanoparticles (Fig. 2c), and the atomic ratio was almost 1:1. All these results demonstrated the successful preparation of zwitterionic SiO2 nanoparticles

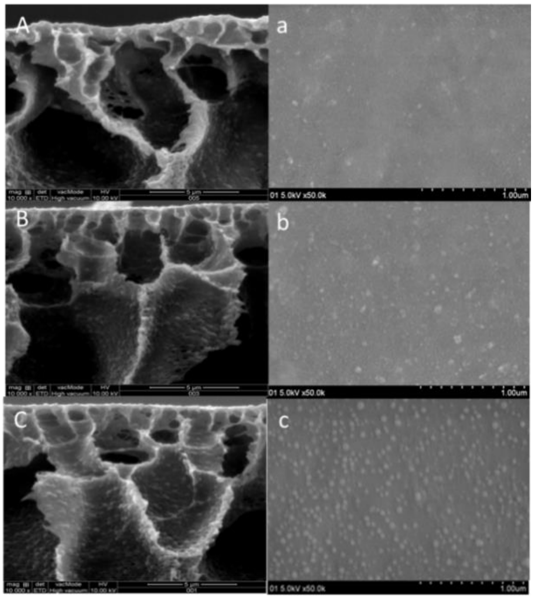

Fig. 3 SEM images of PVDF and hybrid PVDF membranes (A,a: PVDF, B,b: ZP-2%, C,c: ZP-5%)

The effect of zwitterionic SiO_2 nanoparticles addition for the morphology of PVDF ultrafiltration membrane were characterized by SEM as shown in Fig. 3, As be seen from the cross sectional view (A, B, C) of the PVDF, ZP-2% and ZP-5% membranes, PVDF and hybrid membranes exhibited conventional asymmetric structure with cortex layer and finger hole of polymer membranes, for the cross-sectional view images, the massive particle agglomeration problem were not observed clearly. In addition, from the surface images (a, b, c) of the PVDF, ZP-2% and ZP-5% membranes, a large number of nanoparticles were observed on the ZP-2% and ZP-5% membranes surface, and the observed number of nanoparticles on the hybrid membrane surface increased with the addition amount of zwitterionic SiO_2 nanoparticles in PVDF membrane. It can be seen that the plentiful nanoparticles are uniformly distributed on the hybrid membrane surface without obvious agglomeration phenomenon from the surface SEM image of ZP-5%. All these SEM results showed that the modified nanoparticles could be dispersed evenly in the hybrid PVDF membranes, which provided foundation for improving the hydrophilicity of hybrid PVDF membranes. In addition, it should be noted that there will be significant agglomeration of nanoparticles when the added amount was more than 15% in the experiment.

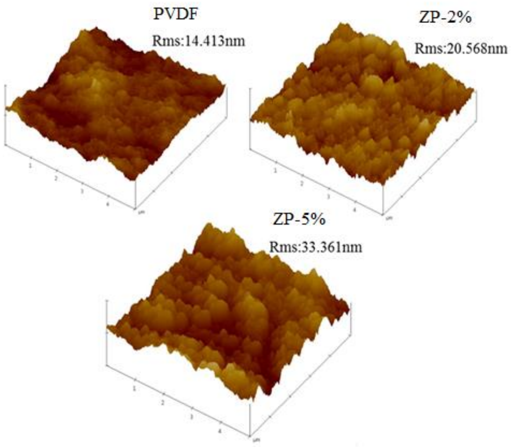

Fig. 4 The AFM images of PVDF and hybrid PVDF membranes

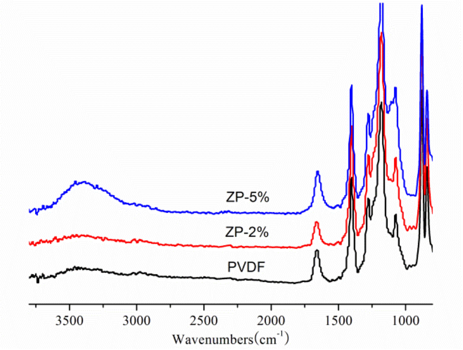

Fig. 5 ATR-FTIR spectra of PVDF and hybrid PVDF membranes

To further investigate the hybird influence of ZP for the morphology of PVDF membranes, surface roughness of hybrid PVDF membranes were measured by AFM. As shown in Fig 4, obiviously, the membrane surface roughness of nanoparticles changed significantly after zwitterionic SiO_2 nanoparticles being added into PVDF membranes, and the different addition amount effect the surface roughness of hybrid membrane. When the addition amount of zwitterionic SiO_2 nanoparticles was 5%, the highest roughness value of 33.361nm was obtained.

To examine the impact of nanoparticles added for the interface physical and chemical properties of PVDF membranes, ATR-FTIR was used to characterize the surface composition of hybrid PVDF membranes, Fig. 5 showed the ATR-FTIR spectra of PVDF, ZP-2% and ZP-5% membranes, It could be confirmed that there were no significant difference between the spectra of PVDF and ZP-2%, indicating that the membrane surface composition did not change significantly, with the added amount of zwitterionic SiO_2 nanoparticles less than 2%. However, there was apparent peak at 1103cm^{-1} which was contributed to the Si-O bond characteristic absorption peak on the spectra of ZP-5% membrane, and the peak intensity of hydroxyl group at 3400 cm^{-1} increased significantly, indicating the presence of SiO_2 nanoparticles on the membrane surface, there are numerous of hydroxyl, amino and carboxyl groups on the zwitterionic SiO_2 nanoparticles surface which increased the number of polar groups on membrane surface thereby enhancing the 3400 cm^{-1} peak intensity. Increased polarity groups such as hydroxy, carboxy and amino groups were beneficial to improve the hydrophilic performance of PVDF membrane.

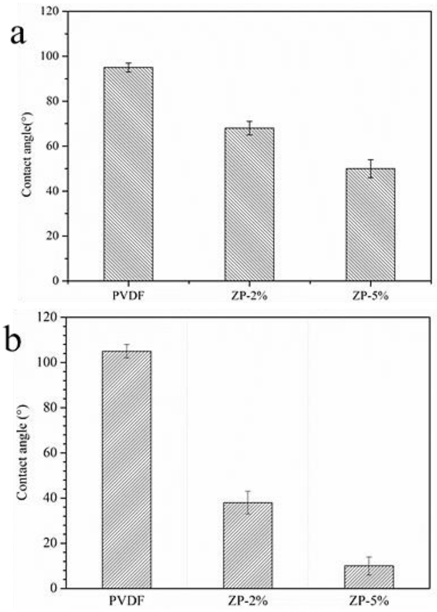

Fig. 6 Water contact angle of membrane surface (a) and mimetic membrane matrix (b)

Spin coating was one of the most used methods for evenly spreading single polymer or nanoparticle onto the substrate.[35] Thus, hybrid membrane solution could be spread over the substrate uniformly, then thermosetting directly to suppress the generation of skin layer on membrane surface, and mimetic porous structure of membrane matrix was obtained. As can be seen from Fig. 6, the contact angle of membrane surface and mimetic matrix for hybrid PVDF membranes decreased distinctly, the surface contact angle of ZP-5% membrane decreased from 95 ° to about 50 °, and the contact angle of mimetic membrane matrix lowered to 10 °. Obviously, the presence of zwitterionic SiO_2 nanoparticles in hybrid PVDF membranes improved the wetting ability of entire hydrophobic PVDF membrane, especially the interior of the membrane.

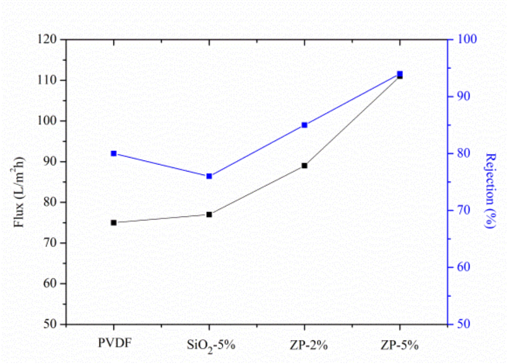

Fig. 7 Separation performance of PVDF and hybrid PVDF membranes

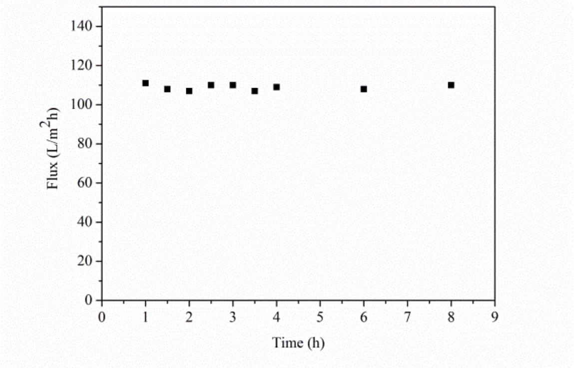

Fig. 8 Time depend water flux of ZP-5% membrane at 0.1MPa

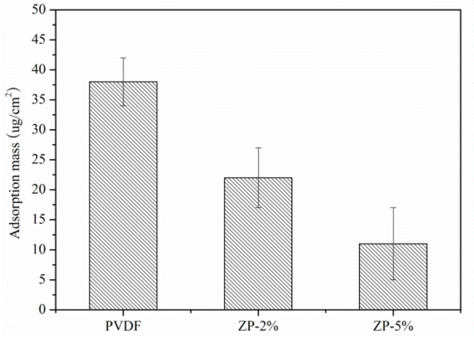

Fig. 9 Static adsorption of BSA solution of PVDF and hybrid PVDF membranes

The effects of added zwitterionic nanoparticles for physicochemical properties and microstructure were detailed characterized above, in addition, the effects on the separation performance of PVDF membrane were also studied, Fig. 7 showed the separation performance of PVDF membranes before and after hybrid modification. The separation data showed that the pure water flux of ZP-5% hybrid membrane increased from 75 L/m^{2}h to 113 L/m^{2}h compared with pristine PVDF membrane with the increased ratio of 50%, while its rejection rate was also improved from 80% to 93%, suggesting that the addition of hydrophilic nanoparticles improved the separation efficiency of hydrophobic PVDF membrane and broken the traditional trade-off phenomenon between flux and rejection ratio, and the exhibited separation performance of ZP-2% and ZP-5% membranes were all better than the SiO_2-5% membrane. The added ZP particles improved the hydrophilicity of PVDF membranes, leading to the improvement of water flux and sieve capacity to BSA molecules of PVDF hybrid membranes, BSA molecules were repulsed from hydrophilic surface and the water molecules were preferentially adsorbed and come though the hydrophilic membrane, thus improving the separation of flux and rejection. The stability of ZP in membrane matrix was tested though the long-time test in water environment, as shown in Fig. 8, there was no obvious change in water flux after the test of eight hours, indicating the nanoparticles would not be lost in the separation process.

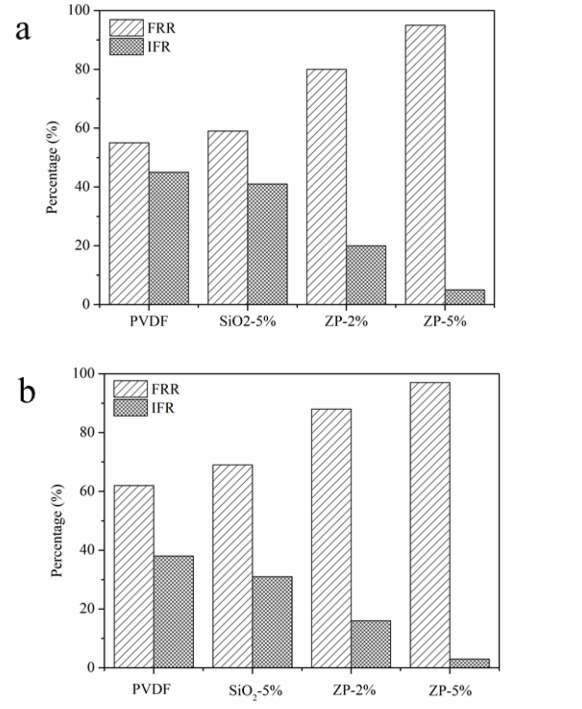

Fig. 10 Dynamic fouling performance of PVDF and hybrid PVDF membranes (a: BSA solution, b: HA solution)

Antifouling ability of PVDF hybrid ultrafiltration membranes were evaluated by static fouling adsorption and dynamic fouling with common pollutants, static fouling test results of PVDF membrane before and after hybridization were shown in Fig. 9, from the adsorption mass date of BSA solution on PVDF membranes, it can be seen that BSA adsorption of per unit area on ZP-modified PVDF hybrid membranes decreased rapidly and BSA adsorption mass of ZP-5% membrane was only 10 μg/cm^2, suggesting the improvement of surface wetting ability by zwitterionic nanoparticles enhanced its anti-adsorption capacity for hydrophobic pollutants and conduced to improve the static antifouling ability of PVDF membranes. When the antifouling characteristics of zwitterionic nanomaterials on the membrane surface were active used, Non-specific adsorption of hydrophobic contaminants in the material interface would be suppressed.

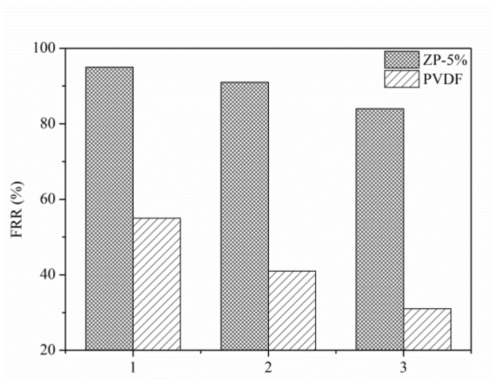

Fig. 11 Repeat antifouling performance of PVDF and ZP-5% membrane (3 times, BSA as pollutant).

Fig. 10 showed the results of dynamic fouling test of PVDF and hybrid PVDF membranes with BSA and HA as two kinds of typical pollutants. As can be seen from FRR value and IFR values of BSA dynamic fouling in the Fig.10 (a), the FRR values of ZP-2% and ZP-5% hybrid membranes were 81% and 95%, respectively, were significantly higher than the 55% corresponding to the FRR value of pristine PVDF membrane, the more zwitterionic nanoparticles added, the greater FRR values obtained, and the relative IFR values were smaller, indicating the added zwitterionic SiO_2 nanoparticles were beneficial to reduce the loss of water flux caused by irreversible fouling, and hydrophobic pollutants were more likely to be cleaned so that the accumulation of pollutants on pore surface and membrane surface decreased rapidly. The dynamic fouling result of HA solution was listed in Fig.10 (b), it could be known that the result showed the same trend with BSA fouling test and the FRR value increased to 97% of ZP-5% membrane, the irreversible fouling caused by HA were significantly reduced due to the addition of zwitterionic SiO_2 nanoparticles, hybrid PVDF membranes exhibited higher water flux recovery rate and reusability. It was worth noting that the FRR values of SiO_2-5% membrane were not significantly improved, which should be due to the reunification problem of nanoparticles. Fig.11 showed long-term antifouling effect of ZP, even after the dynamic fouling test of 3 times, the FRR value of ZP-5% membrane was no less than 85%, showing the hybrid membrane would have a longer service life.

All these results demonstrated that hybrid of PVDF membrane with zwitterionic nanoparticles enhanced the antifouling performance of hydrophobic PVDF membranes and improved the membrane separation efficiency though reducing the irreversible fouling caused by irremovable pollutants. The addition of zwitterionic SiO_2 nanoparticle fabricated hydrophilic membrane surface and membrane matrix, based on the hydrophilic and antifouling properties of modified zwitterionic SiO_2 nanoparticle, the wettability of entire hydrophobic PVDF membrane was significantly improved, water molecules were preferentially adsorbed on the hydrophilic membrane surface and pore surface, so that anchored effect of pollutants on hydrophobic porous membrane was weakened, leading to be washed away easily.

4. Conclusions

Zwitterionic SiO_2 nanoparticles were blended with PVDF to prepare hybrid PVDF ultrafiltration membrane, the distribution of zwitterionic SiO_2 nanoparticles was extremely uniform in hybrid PVDF ultrafiltration membranes, indicating the reunion feature of nanoparticles was effectively inhibited. Furthermore, the surface wetting properties and separation performance of PVDF membranes were all significantly improved due to the addition of zwitterionic SiO_2 nanoparticles. Meanwhile, the blended zwitterionic SiO_2 nanoparticles significantly enhanced the antifouling performance of PVDF membranes. BSA adsorption mass of ZP-5% membrane decreased to 10 $\mu g/cm^2$ in static fouling test and the FRR values of secondary water flux were no less than 95% for the dynamic fouling test using BSA and HA pollutants, the excellent antifouling performance of PVDF membranes results indicating that the hybrid modification with anchored zwitterionic material is an effective and convenient method to enhance the fouling resistance of hydrophobic PVDF membrane.

Acknowledgements

This work is supported by grants from the National Science Foundation of China (No.21174027), Program for New Century Excellent Talents in University (No.NCET-12-0827). Program of Introducing Talents of Discipline to Universities (No. 111-2-04) and Innovation Funds for PhD Students of Donghua University (CUSF-DH-D-2015027).

References

1 M. A. Shannon, P. W. Bohn, M. Elimelech, J. G. Georgiadis, B. J. Marinas and A. M. Mayes, Nature, 2008, 452, 301−310.
2 W. B. Yang, N. Cicek and J. Ilg, J. Membr. Sci., 2006, 270, 201–211.
3 K. J. Howe and M. M. Clark, Environ. Sci. Technol., 2002, 36, 3571–3576.
4 P. Le-Clech, V. Chen, and T. A. Fane, J. Membr. Sci., 2006, 284, 17–53.
5 S.F. Chen, L.Y. Li, C. Zhao and J. Zheng, Polymer, 2010, 51, 5283–5293.
6 J. B. Schlenoff, Langmuir, 2014, 30, 9625-9636.
7 I. Banerjee, R. C. Pangule and R. S. Kane, Adv. Mater., 2011, 23, 690−718.
8 M. Kobayashi, Y. Terayama, H. Yamaguchi, M. Terada, D. Murakami, K. Ishihara and A. Takahara, Langmuir, 2012, 28, 7212−7222.
9 D. Rana and T. Matsuura, Chem. Rev., 2010, 110, 2448−2471.
10 I. Banerjee, R. C. Pangule and R. S. Kane, Adv. Mater.,

2011, 23, 690−718.
11 M. Kobayashi, Y. Terayama, M. Kikuchi and A. Takahara, Soft Mater., 2013, 9, 5138−5148.
12 S. F. Chen, J. Zheng, L. Y. Li and S. Y. Jiang, J. Am. Chem. Soc. 2005, 127, 14473−14478.
13 J. Ladd, Z. Zhang, S. Chen, J. C. Hower and S. Jiang, Biomacromolecules, 2008, 9, 1357−1361.
14 Z. G. Estephan, P. S. Schlenoff and J. B. Schlenoff, Langmuir, 2011, 27, 6794−6800.
15 Y. F. Yang, Y. Li, Q. L. Li, L. S. Wan and Z. K. Xu, J. Membr. Sci., 2010, 362, 255−264.
16 J. H. Kuang and P. B. Messersmith, Langmuir, 2012, 28, 7258−7266.
17 A. B. Lowe, N. C. Billingham and S. P. Armes, Macromolecules, 1999, 32, 2141−2148.
18 J. D. Grooth, D. M. Reurink,; J. Ploegmakers, W. M. de Vos and K. Nijmeijer, ACS Appl. Mater. Interfaces., 2014, 6, 17009–17017.
19 Y. F. Zhao, L.P. Zhu, Z. Yi, B.K. Zhu and Y.Y. Xu, J. Membr. Sci., 2013, 440, 40–47.
20 A .K. Singh, P. Singh, S. Mishra and V. K. Shahi, J. Mater. Chem., 2012, 22, 1834– 1844.
21 J. Zhang, Z. Xu, W. Mai, C. Min, B. Zhou, M. Shan, Y. Li, C. Yang, Z. Wang and X. Qian, J. Mater. Chem. A, 2013, 1, 3101–3111.
22 F. Liu, M. R. Abed and K. Li, J. Membr. Sci., 2011, 366, 97−103.
23 Y. H. Yin, T. Xu, X. H. Shen, H. Wu and Z.Y. Jiang, J. Membr. Sci., 2014, 469, 355−363.
24 H. Wu, J. Mansouri and V. Chen. J. Membr. Sci., 2013, 433, 135–151.
25 A. S. Gorzalski and O.Coronell, J. Membr. Sci., 2014, 468, 349−359.
26 S. Hassanajili, M. Khademi and P. Keshavarz, J. Membr. Sci., 2014, 453, 369−383.
27 Q.Y. Wang, Z.W. Wang, J. Zhang, J. Wang and Z.C. Wu, RSC Adv., 2014, 4, 43590−43598.
28 F.M. Jin, W. Lv, C. Zhang, Z.J. Li, R.X. Su, W. Qi, Q. H. Yang and Z.M. He, RSC Adv., 2013, 3, 21394−21397.
29 A. M. Alswieleh, N. Cheng, I. Canton, B. Ustbas, X. Xue, V. Ladmiral, S. J. Xia, R. E. Ducker, O. E. Zubir, M. L. Cartron, C. N. Hunter, G. J. Leggett and S. P. Armes, J. Am. Chem. Soc., 2014, 136, 9404−9413.
30 A. B. Lowe, and C. L. McCormick, Chem. Rev., 2002, 102, 4177−4189.
31 C. Blaszykowski, S. Sheikh and M. Thompson, Chem. Soc. Rev., 2012, 41, 5599−5612.
32 Y. Liu, C. Ma, S. F. Wang, H. X. Guo, B. H. Zhang, L. Zhang, K. L. Gu and J. Y. Gu, RSC Adv., 2015, 5, 21316−21325.
33 B. P. Tripathi, N. C. Dubey, S. Choudhury and M. Stamm, J. Mater. Chem., 2012, 22, 19981−19992.
34 H. Jiang and F.J. Xu, Chem. Soc. Rev., 2013, 42, 3394−3426.
35 C. Sanchez, P. Belleville, M. Popall and L. Nicole, Chem. Soc. Rev., 2011, 40, 696−753.