Editors

Editor-in-Chief:
Michel A. Aegerter

Co-Editors:
Florence Babonneau, Sorbonne Université, France

Yunfa Chen, Chinese Academy of Sciences, China

Andrei Jitianu, Lehman College-City University of New York, USA

Kazuki Nakanishi, Nagoya University, Japan

Special Section Editor:
John Bartlett, University of the Sunshine Coast, Australia

Assistant Editors:
Xiaobo Chen, University of Missouri – Kansas City, USA

Paolo Falcaro, Graz University of Technology, Austria

Julian R. Jones, Imperial College London, United Kingdom

Luca Malfatti, University of Sassari, Italy

Alessandro Martucci, Università di Padova, Italy

Yasuaki Tokudome, Osaka Prefecture University, Japan

Editorial Board:

Rui M. Almeida, Instituto Superior Técnico, Portugal; David Avnir, Hebrew University of Jerusalem, Israel; Beyong-Soo Bae, Korea Advanced Institute of Science and Technology (KAIST), Korea; Luis D. Carlos, University of Aveiro, Portugal; Alicia Durán, Instituto de Cerámica y Vidrio (CSIC), Spain; Luis M. Esquivias, Universidad de Sevilla, Spain; Xianping Fan, Zhejiang University, China; Massimo Guglielmi, University of Padova, Italy; Nicola K. Huesing, University of Salzburg, Austria; Galo Soler-Illia, Comision Nacional de Energia Atomica (CNEA), Argentina; Plinio Innocenzi, Universita degli Studi di Sassari, Italy; Vadim G. Kessler, Swedish University of Agricultural Sciences, Sweden; Hiromistu Kozuka, Kansai University, Japan; David Levy, Instituto de Ciencia de Materiales de Madrid (ICMM, CSIC), Spain; Atsunori Matsuda, Toyohashi University of Technology, Japan; Jean-Marie Nedelec, SIGMA Clermont, France; Michael Popall, Fraunhofer-Institut für Silicatforschung, Germany; Michel Prassas, Corning Europe Inc., France; Sidney J. L. Ribeiro, Institute of Chemistry-UNESP, Brazil; Celso V. Santilli, Institute of Chemistry-UNESP, Brazil; Jun Shen, Tongji University, China; Leon L. Shaw, Illinois Institute of Technology, USA; Kiyoharu Tadanaga, Hokkaido University, Japan; Marlies K. Van Bael, University of Hasselt & IMEC, Belgium; Maria M. Zaharescu, Institute of Physical Chemistry, Romania

For authors
Volume 96, issue 3, December 2020

22 articles in this issue

Role of starch in the ceramic powder synthesis: a review
Joanna Mastalska-Poplawksa, Marek Sikora ... Zuzanna Góral
Review Paper: Nano-structured materials (particles, fibers, colloids, composites, etc.)
Published: 10 September 2020 | Pages: 511 - 520

Research on magnetic and microwave absorbing properties of Co₂Y ferrite fabricated by sol-gel process
Yu He, Shunkang Pan & Jingjing Yu
Original Paper: Sol-gel and hybrid materials for dielectric, electronic, magnetic and ferroelectric applications
Published: 02 February 2020 | Pages: 521 - 528

Study on microstructural and electro-optical properties of sol–gel derived pure and Al/Cu-doped ZnO thin films
Fatemeh Dabir, Hamid Esfahani ... Zahra Khodadadi
Original Paper: Sol-gel and hybrid materials for optical, photonic and optoelectronic applications
Published: 07 March 2020 | Pages: 529 - 538

The effect of MgO addition on the properties of alumina-based ceramic cores prepared via sol–gel process
Shaghayegh Shabani, Rahim Naghizadeh ... Farhad Golestanifard
Original Paper: Sol-gel, hybrids and solution chemistries
Published: 16 May 2020 | Pages: 539 - 549

Reverse microemulsion synthesis of mixed α and β phase NaYF₄:Yb,Er nanoparticles: calcination induced phase formation, morphology, and upconversion emission
M. Gunaseelan, S. Yamini ... J. Senthilvelan
Original Paper: Sol-gel and hybrid materials for optical, photonic and optoelectronic applications
Published: 25 June 2020 | Pages: 550 - 563

Sol–gel synthesis of iodosodalite precursors and subsequent consolidation with a glass binder made from oxides and sol–gel routes
Jared O. Kroll, Brian J. Riley ... Jacob A. Peterson
Original Paper: Sol–gel and hybrid materials for energy, environment and building applications
Published: 28 June 2020 | Pages: 564 - 575

Fabrication of a sol–gel polishing tool for green manufacturing of the seal stone
Qiufa Luo, Jing Lu ... Jian Wang
Original Paper: Sol-gel and hybrid materials for energy, environment and building applications
Published: 09 July 2020 | Pages: 576 - 588
Effects of NaOH amount on condensation mechanism to form aluminosilicate, case study of geopolymer gel synthesized via sol–gel method

Xu Chen & Paramita Mondal
Original Paper: Sol–gel and hybrid materials for energy, environment and building applications
Published: 12 July 2020 | Pages: 589 - 603

A comparative study of microstructure, magnetic, and electromagnetic properties of Zn₂W hexaferrite prepared by sol–gel and solid-state reaction methods

Lining Fan, Hui Zheng ... Yang Zhang
Original Paper: Sol–gel and hybrid materials for dielectric, electronic, magnetic and ferroelectric applications
Published: 16 July 2020 | Pages: 604 - 613

Fast responsive thermally stable silica microspheres for sensing evaluation: sol–gel approach

Shumaila Islam, Hazri Bakhtiar ... Shahzad Naseem
Original Paper: Sol–gel, hybrids and solution chemistries
Published: 16 July 2020 | Pages: 614 - 626

Microencapsulated phase change materials composites Al₂O₃–SiO₂ aerogel and the thermal regulation properties

Yiting Zhang, Jing Li ... Zhong Chen
Original Paper: Sol–gel and hybrid materials for energy, environment and building applications
Published: 17 July 2020 | Pages: 627 - 635

Preparation of fluoroalkyl end-capped vinyltrimethoxysilane oligomer/micro-sized silica composites possessing superoleophilic/superhydrophobic characteristic: application to selective removal of aromatic compounds from aqueous methanol solution by using these composites

Hideo Sawada, Masaya Chiba ... Jun-ichi Suzuki
Original Paper: Sol–gel and hybrid materials with surface modification for applications
Published: 17 July 2020 | Pages: 636 - 648

Sol–gel synthesis and structural characterization of band gap engineered ferroelectric perovskite oxide potassium sodium barium nickel niobate

Yunxin Li, Shulan Zhang, Shun Song
friendly sol–gel method
Nurul Pratiwi, Zulhadji ... Diana Vanda Wella
Original Paper: Sol–gel and hybrid materials with surface modification for applications | Published: 21 August 2020 | Pages: 669 - 678

Investigating the mechanism of uniform Ag@SiO₂ core-shell nanostructures synthesis by a one-pot sol–gel method
Mohammad Jamir Ahemad & Yeon-Tae Yu
Original Paper: Sol-gel, hybrids and solution chemistries | Published: 26 August 2020 | Pages: 679 - 689

Effect of the addition of calcium salts on the structure and anticorrosion properties of siloxane-poly(hydroxyethyl methacrylate) hybrid coating applied on Ti-6Al-4V alloy
Larissa de Lima Almeida, Deidyane Wityla Félix da Silva Ferreira ... Victor Hugo Vitorino Sarmento
Original Paper: Sol-gel and hybrid materials with surface modification for applications | Published: 29 August 2020 | Pages: 690 - 701

Investigation of morphological and electrical properties of RTA-processed TiO₂ for memristor application
S. Roy, S. P. Ghosh ... I. P. Kar
Original Paper: Sol-gel and hybrid materials for dielectric, electronic, magnetic and ferroelectric applications | Published: 03 September 2020 | Pages: 702 - 717

Study of structural, optical, and dielectric properties of sol–gel derived ZnFe₂O₄–Al₂O₃ composite nanoparticles
Ashwini S. Varpe & Mrinalini D. Deshpande
Self-cleaning material based on superhydrophobic coatings through an environmentally friendly sol–gel method

Nurul Pratiwi, Zulhadjri, Syukri Arief, Admi & Diana Vanda Wellia
Your article is protected by copyright and all rights are held exclusively by Springer Science+Business Media, LLC, part of Springer Nature. This e-offprint is for personal use only and shall not be self-archived in electronic repositories. If you wish to self-archive your article, please use the accepted manuscript version for posting on your own website. You may further deposit the accepted manuscript version in any repository, provided it is only made publicly available 12 months after official publication or later and provided acknowledgement is given to the original source of publication and a link is inserted to the published article on Springer’s website. The link must be accompanied by the following text: “The final publication is available at link.springer.com”.

Springer
Self-cleaning material based on superhydrophobic coatings through an environmentally friendly sol–gel method

Nurul Pratiwi1 · Zulhadji1 · Syukri Arief1 · Admi1 · Diana Vanda Wellia1

Received: 27 April 2020 / Accepted: 12 August 2020 / Published online: 21 August 2020
© Springer Science+Business Media, LLC, part of Springer Nature 2020

Abstract
Recently, there has been increasing interest in developing artificial superhydrophobic surfaces, especially in the field of self-cleaning application. However, the poor robustness and high-cost preparation of these surfaces have always been some issues for their industrial development. Herein, we describe an environmentally friendly way to prepare stable, robust, and transparent superhydrophobic coatings through the deposition of a rough substructure of TiO2 film followed by chemical modification using octadecyltrichlorosilane (OTS), a fluorine-free organic silane. The as-prepared coatings exhibited a great superhydrophobic property and ultralow adhesion (with a static water contact angle of 158 ± 2° and sliding angle of 4 ± 1°). It was found that the superhydrophobic coatings can still maintain good performance after UV irradiation, chemical immersion, and physical abrasion. More importantly, the coated surfaces showed an excellent self-cleaning ability against dirt particles after rinsed with water droplets.

Graphical Abstract

Keywords Superhydrophobic · Octadecyltrichlorosilane · TiO2 · Transparent glass · Self-cleaning
Highlights

- Environmentally friendly and low-cost method to prepare superhydrophobic coatings using TiO$_2$ thin film and monolayer octadecyltrichlorosilane (OTS).
- High transparency of the as-prepared superhydrophobic coatings.
- The superhydrophobic coatings exhibited good chemical and mechanical stability.
- The surface of superhydrophobic glass showed an excellent self-cleaning property.

1 Introduction

The super-wettability feature on the surface and interface of solid material has become one of the essential aspects of surface chemistry development [1]. Over the last decades, superhydrophobic surfaces, an excellent nonwetting systems, have gained remarkable attention due to their tremendous applications in self-cleaning [2], antifouling [3], heat exchangers [4], oil-water separation [5], smart sensing [6], and thermal insulation [7]. Inspired by nature, superhydrophobic surfaces, which have static water contact angle (CA) $>$ 150° and sliding angle (SA) $<$ 10°, are constructed by two main factors, hierarchically micro-/nanoscale surface structure (for trapping air) and surface chemical compositions (for lowering the surface energy) [8].

Various methods have been used to prepare artificial superhydrophobic coatings, such as chemical vapor deposition [9], photolithography [10, 11], RF magnetron sputtering [12], and electrospraying [13]. Even though superhydrophobic surfaces could be produced, most of all these reported methods still involve complicated procedures, high energy consumption and difficulty in scaling-up [14]. Therefore, a low-cost and simple method for preparing superhydrophobic coatings, such as wet-chemical methods have been considered as an alternative method [15].

The sol–gel method assisted with the dip-/spray-/spin-coating technique was one of the favored method to create the surface roughness by incorporating the micro-/nano of inorganic particles [16]. Following by chemical surface modification with low surface energy materials, the artificial superhydrophobic surfaces could be successfully achieved [17]. There are several studies that have been reported on developing superhydrophobic coating through this combination method. Kong et al. have prepared superhydrophobic coating on glass substrate by fabricated a micro-/nanos-structured TiO$_2$ film via the sol–gel dip-coating method and then modified by fluoroalkylsilane molecules [18]. The as-prepared coatings not only showed a high static water CA up to 165.6° but also enhanced the reversible wettability conversion between hydrophobicity and hydrophilicity. However, the transparency and stability of coatings were not clearly mentioned. As an improvement to avoid the use of fluorine compound, Zhang et al. prepared superhydrophobic SiO$_2$-based coating onto the glass substrate by a sol–gel spin-coating technique at low temperature and then followed by surface modification of trimethylchlorosilane [19]. As the results, the obtained coatings exhibited high transparency, good robustness, and excellent antiwetting behavior with a static water CA of 164 ± 2°. However, the use of a large amount of organic solvent in this work may potentially be a harmful pollutant for the environmental.

Based on all of these drawbacks, the fabrication of superhydrophobic coatings is quite limited for some reasons: first, methods that widely used to fabricate superhydrophobic coatings are costly and tricky [20]. Second, some chemicals that involved in the fabrication of superhydrophobic coatings such as fluorinated compounds are very hazardous due to their nondegradable and high toxic properties [21]. Third, the high surface roughness that is needed to give the superhydrophobic coatings is always contradictory with the transparency properties [22]. Forth, the robustness and stability of artificial superhydrophobic coatings are still limited in the practical application [23]. Therefore, the development of a coating with sustainable superhydrophobicity and good coating integrity through an environmentally safe method is highly desirable.

In our previous work, we discussed the optimum parameters in fabricating hydrophobic coatings on the glass surface by using a facile and environmentally friendly method [24]. The high surface roughness of TiO$_2$ film was obtained through a simple peroxo sol–gel dip-coating method. The effect of wettability with different concentrations of silane compounds was also investigated. Even though showing a high transparency, the static water CA was found of 146°, which was categorized as “ultra-hydrophobic.” In this study, an environmentally friendly and cost-effective method to prepare stable, robust, and transparent superhydrophobic coatings were developed to improve the previous work. The coating was prepared by chemical deposition of TiO$_2$ films through a simple peroxo sol–gel method on glass substrates and its subsequent dipping modification with octadecyltrichlorosilane (OTS), fluorine-free compounds. This approach is considered as a green technology since the use of water solvent and neutral pH without any hazardous chemicals. Moreover, this paper describes the morphological, mechanical, and chemical stability as well as the self-cleaning ability of the as-prepared coatings.
2 Materials and methods

2.1 Materials

Titanium tetrachloride (TiCl₄ 0.09 M in 20% HCl), ammonium hydroxide (NH₄H₂O₂ 25%), and OTS (C₁₈H₃₇Cl₃Si ≥ 90%) were purchased from Aldrich. Hydrogen peroxide (H₂O₂ 30%) and anhydrous ethanol (C₂H₆O ≥ 99.5%) were purchased from Merck. All chemicals were analytical reagent grade and used without any further purification. The sodalime glass slides (CAT. No. 7101) with a thickness of 1 mm were purchased from Sail brand.

2.2 Preparation of TiO₂ sol

TiO₂ sol was synthesized with the typical peroxo sol–gel method [24, 25]. At the temperature condition of 5 °C, the drop-wise of 36 mL TiCl₄ was dissolved in 300 mL deionized water under strong magnetic stirring for 30 min and then the solution was neutralized by the drop-wise addition of ammonia solution. This solution was kept stirring vigorously for 24 h to obtain the fine white precipitates. These precipitates were washed repeatedly with deionized water until no Cl⁻ ions were detected. Finally, the obtained precipitates were dissolved in 80 mL deionized water at ambient temperature. Then 28 mL H₂O₂ was added drop-wise into the mixture under constant stirring for 4 h to form an aqueous transparent yellowish peroxotitanium complex (PTC) solutions.

2.3 Fabrication of TiO₂ films on the glass substrate

The cleaning treatment procedures of glass slide were adopted from the previous work [24, 25]. The precleaned glass slide was dipped in PTC solutions for 30 s and was withdrawn using a dip-coater with a speed rate of 0.1 cm/s. Then, the coated glass was dried in vacuum at 80 °C for 5 min. This coating procedure was repeated five times. Finally, the coated glass was cured at 120 °C in vacuum for 1 h.

2.4 Preparation of superhydrophobic surfaces

The TiO₂ coated glass was placed into the OTS/ethanol solution (2%, v/v) at room temperature for 6 h. The obtained coatings were rinsed with ethanol and dried at 60 °C for 3 h in a vacuum oven. Monolayer OTS-coated glass was prepared as well for comparison.

2.5 Characterization

Surface morphologies and topographies of the coatings were observed by field emission scanning electron microscopy (FESEM, FEI Inspect F50) and atomic force microscopy (AFM, Bruker nanoscan 9). The chemical compositions of coated and uncoated glass slides were investigated by Fourier-transform infrared spectroscopy (FTIR, PerkinElmer) and energy dispersive spectroscopy (EDS, EDAX Apollo X). Water CA and SA were measured at ambient condition by CA analyzer (SEO Phoenix 300) with 5 μL of water droplets. The CA measurements were taken at five different positions, and an average of total measurement was used to determine the final CA value. The SA value was measured by tilting the sample until the water droplet roll-off. An average of five times measurement on each sample was applied as the final SA value.

2.6 Mechanical test

The robustness of the as-prepared superhydrophobic coatings was investigated by the conventional abrasion tests. The as-prepared coated glass slide was placed face down on standard sandpaper (1500 mesh), and the pressure was set up by placing a weight (100 g) on top of it. Then, the average of CA and SA values were measured after dragging the coated glass slide horizontally with the speed rate of 0.16 cm/s for 100 cm.

2.7 Chemical test

The CA and SA values of the as-prepared coated glass slide were measured after varying the pH value of the drops with a pH range of 1–13. Moreover, to find out the durability of the coated glass slide to extreme environments, the CA and SA values of this coating were measured after being dipped in neutral (pH 7), acid (H₂SO₄, pH 4), and base (NaOH, pH 10) solutions for 20 h.

2.8 UV exposure test

The coated glass slide was placed horizontally in a close reactor with UV A blacklight blue (λ = 352 nm, Camag). The distance between the substrate and the UV lamp was set up for 13 cm. Then, these coating was irradiated by UV light for 5 h. At interval 60 min, the average CA and SA values of this irradiated coating were measured.

2.9 Self-cleaning test

Carborundum particles (with sizes in the range of 37–44 μm) were used as a model pollutant. The procedure of the self-cleaning test was carried out by setting up the coated glass slide in a clean petri dish at an angle of ~10°, and the carborundum particles were placed on top of it. Then, the cleaning process was evaluated after the water droplets were dropped onto the polluted coated glass slide. The uncoated glass was used as a comparison.


3 Results and discussion

3.1 Morphology, composition, and wettability of as-prepared superhydrophobic coatings

The schematic illustration of the superhydrophobic coatings fabrication on a glass substrate was shown in Fig. 1. Herein, the TiO₂ films were used to construct the sub-rough structure on the hydrophilic glass surface. It is found that the chemical interaction occurs after the hydroxyl group =Ti–OH on TiO₂ film and ≡Si–OH on the pretreated glass slide condense, forming cross-linked chains of ≡Si–O–Ti≡ [24, 25]. As a result, the TiO₂ films were coated on the glass surface with a good surface roughness. However, in order to decrease the surface energy on the roughened surface of TiO₂-coated glass, the OTS monolayer was grafted on it. The main reaction occurs when the head of OTS (–SiCl₃) physisorbs and then react with the adsorbed water layer on the surface of the TiO₂-coated glass to form a silanol group (–Si(OH)₃). After that, this hydrolyzed head of OTS (–Si(OH)₃) condenses with OH-groups on the surface of TiO₂ films and then forms a strong chemical interaction of ≡Si–O–Ti≡. Consequently, the OTS molecules are firmly grafted on the surface of TiO₂-coated glass [26, 27]. As the condensation reaction occurs among the neighboring OTS molecules, a densely long-chain alkyl groups (–C₁₈H₃₇) of OTS tail are arranged and finally decreased the surface energy of the as-prepared coated glass [24, 28]. Through this fabrication mechanism, the as-prepared coated glass not only showed superhydrophobic property but also exhibited transparent and stable coatings.

Figure 2 presents the shapes of static waterdrops on the uncoated, TiO₂-coated, OTS-coated, and superhydrophobic glass, respectively. As shown in Fig. 2a and b, both the uncoated glass and the TiO₂-coated glass exhibit hydrophilic property with CAs < 90° which are mostly attributed by surface roughness and high surface energy from these surfaces [29, 30]. Contradictory, the glass slide that coated with OTS monolayer shows the hydrophobic property with CA of 133 ± 3° (Fig. 2c) due to the nonpolar long alkyl chain of OTS that arranged in the layer to lower the surface energy of the glass slide [31]. However, after the TiO₂-coated glass slide was subsequently treated by OTS, the
glass slide exhibited a good superhydrophobic property with CA as high as $158 \pm 2^\circ$ (Fig. 2d). Here, through the combination of the roughened surface of TiO$_2$ film and the low-surface-energy of OTS, the superhydrophobicity of the as-prepared coated glass surface can be explained according to Cassie–Baxter model [32]. In this model, it is presumed that the water droplet is situated on solid–vapor–liquid interfaces since there is a large amount of trapped air in the grooves within the surface rough structure forming a low adhesion composite surface and significantly causes an
increase of CA as well as a decrease of SA [15, 33, 34]. Thus, a high quality of superhydrophobic surface could be successfully fabricated [14, 15].

As mentioned before, one of the most important factors needed in fabricating the super-wetting system is high surface roughness. Therefore, the surface morphologies and topographies of the as-prepared coating were further studied. Figure 3 shows the FESEM images of uncoated and superhydrophobic glass. It can be seen that the uncoated glass exhibits smooth surface (Fig. 3a), whereas the superhydrophobic glass surfaces are covered by uneven hierarchical structures, which is attributed to the deposition of TiO2 and OTS layer (Fig. 3b). The topographies of this coated glass were further confirmed by AFM analysis (Fig. 3c, d). It is found that the mean roughness \( R_a \) of uncoated glass, and superhydrophobic glass is 2 and 4 nm, respectively.

Figure 4 shows the EDX spectra and the regional analysis of superhydrophobic glass. It was observed that the high spectra of Si and C represent the composition of OTS layer. In contrast, the detected low spectra of Ti and O, which is attributed to the existence of TiO2 compounds on the coated glass surface, confirming that the surface of TiO2 film was mostly grafted by OTS monolayer. Furthermore, the signal of Si, Al, Na, Ca, and Mg elements could be also attributed to the dominant elements of sodalime glass [5].

To investigate the interpretation of chemical structure as well as the chemical interactions, the uncoated and superhydrophobic glass were characterized by FTIR. Figure 5 presents the FTIR spectra of the samples in the range of 4000–400 cm\(^{-1}\). As shown in Fig. 5a, it is observed that the absorption peaks at 922 cm\(^{-1}\) is attributed to the Si–O–Si bend vibration on the sodalime glass. However, after the glass consecutively being coated with TiO2 film and OTS (Fig. 5b), this absorption peak is shifted to the 1019 cm\(^{-1}\), which assigning to asymmetric Si–O–Si stretching vibration as a result of the cross-linked interaction of OTS [35, 36] and to the 902 cm\(^{-1}\), which indicating the interaction of Si–O–Ti groups, confirming the covalent interaction between TiO2 layer with OTS layer and/or TiO2 layer with glass substrate [25, 26, 37]. Besides, the characteristic absorption peaks that only could be found on the coated glass (Fig. 5b) are exhibited at 2914 and 2853 cm\(^{-1}\), which respectively assigned to asymmetrical –CH\(_3\)– and symmetrical –CH\(_2\)– strain vibrations confirming the presence of the long-chain alkyl groups of OTS compounds [38].

### 3.2 Mechanical and chemical stability

In order to evaluate the ability of the coated glass in maintaining its superhydrophobic properties against abrasion from the external environment, the abrasion test was carried out by adapting the previously reported analogical procedures [39, 40]. Figure 6 shows the CA and SA of the superhydrophobic glass as a function of the abrasion length.
ranging the pH values of the liquid droplet. The CA of a liquid droplet on the surface of superhydrophobic glass decreases slightly when the liquid droplet at acid point or base point, but still perform the superhydrophobic property (CA > 150°). On the contrary, the SA increases at the acid and base point, with the average SA value <10°. As the superhydrophobic glass perform high CA of the liquid droplet at pH 7, we further investigate the durability of this coating at this condition. Figure 7b shows the CA and SA of the superhydrophobic surface that immersed in water (pH ~7–8) for 5 days. After the superhydrophobic surface was immersed in water, the CA slightly decreased from 158 ± 1° to 150 ± 1°, and the SA slightly increased from 4° to 9 ± 1° in the last day. These results assert that the as-prepared superhydrophobic glass has good durability in maintaining its superhydrophobicity even though in environment with high relative humidity. The CA and SA of the as-prepared superhydrophobic glass as a function of immersion time in the acid (pH 4) and base (pH 10) solution are present in Fig. 7c and d, respectively. It is found that, as the immersion time increase, the CA of superhydrophobic glass gradually decrease to 153 ± 1° and 152 ± 1° after 18 h immersed in acid and base solution, successively. Meanwhile, the SA of superhydrophobic glass slightly increases to 9 ± 1° and 8 ± 1° after respectively immersing in acid and base solution for 18 h. Here, the acid and base solution could initiate a
hydrolysis and condensation reaction with silane compound, which further could damage the interaction between the OTS layer and TiO$_2$ film resulting in a decrease of CA and increase of SA value, gradually [41, 42]. However, the strong interaction between the interlayer of OTS and TiO$_2$ film on the coated glass surface is believed could slow down this devastation. Therefore, it can be concluded that the as-prepared superhydrophobic glass has good long-term stability in a neutral, acidic and alkaline environment.

To further evaluate the stability of the as-prepared coated glass in outdoor applications, the CA and SA of these coatings after UV exposure were measured. In Fig. 8, the CA of the superhydrophobic glass only slightly decrease to 150 ± 2° from 158 ± 1° and SA gradually increase to 8 ± 3° from 3 ± 1° after UV exposure for 300 min, exhibiting a good stability and durability of the coatings. Generally, when exposed to UV light, the TiO$_2$ films could easily trigger the hydrophilic properties by creating the oxygen vacancies, which further could decrease the CA value. However, the obtained results confirm that the presence of TiO$_2$ layers on the surface of coated glass, after being irradiated by UV light, did not have a significant effect on decreasing the CA value. It is due to the presence of oxygen atoms from OTS molecules can gradually replace the oxygen vacancies on TiO$_2$ film to maintain the hydrophobicity [21].

3.3 Self-cleaning ability

Even though self-cleaning properties generally could be performed in both super-wetting system (superhydrophilic and superhydrophobic) by using water, but the cleaning process for both of these surfaces is entirely different. Herein, the self-cleaning ability on the superhydrophobic surface that we fabricated and applied on the glass substrate will be described. Figure 9 shows the self-cleaning simulation of uncoated and superhydrophobic glass. The cleaning process was observed by capturing the ability of water droplets in removing the carborundum particles, which were placed on the glass surface. As shown in Fig. 9a–d, the carborundum particles on the coated glass surface were removed as quickly as the water droplets rolled-off, obtaining a clean surface. In contrary, the carborundum particles on the uncoated glass were quite difficult to remove with the same procedure (Fig. 9e–h). Generally, the cleaning process performed on the superhydrophobic surface was employed by physical interaction between the water droplets and the contaminant that rolling-off from this surface [43]. To make this happen, high surface roughness and low surface energy must be presented. In this work, TiO$_2$ film was used to increase the surface roughness by creating the hierarchical rough structure. This surface roughness traps a large amount of air between its grooves, meanwhile, the OTS layer was used to decrease the surface energy [14, 38]. Together, those features could maintain the water droplet in the solid–vapor–liquid phase forming a high-water CA and a low-water SA. This explained that the two mentioned features on the as-prepared superhydrophobic glass caused the water droplets easily rolling-off over the modified surface without wetting the surface so that contaminants will be easier to remove [38].

4 Conclusions

Superhydrophobic glass has been successfully prepared through an environmentally friendly and low-cost strategy to prepare mechanically robust, UV stable, chemically resistance, and transparent superhydrophobic coatings by depositing TiO$_2$ film on the glass substrate, followed by surface modification of fluorine-free compound of OTS. The coated glass exhibited excellent superhydrophobic property with CA of 158 ± 2° and ultralow adhesion with an SA of 4 ± 1°. The coated glass has been shown to have good mechanical and chemical stability while maintaining its superhydrophobic surface after 80 cm of abrasion against 1500 mesh sandpaper, and after being immersed in acidic (pH 4) and alkaline (pH 10) solutions for 18 h. Moreover, this modified glass substrate has also been shown to have
good durability while maintaining its superhydrophobic surface after soaking in water for 5 days and UV irradiation for 300 min. Importantly, the as-prepared superhydrophobic glass has a good self-cleaning property, which can be employed to clean contaminants (SiC particles) on its surface quickly compared to uncoated glass. The study showed that this feasible strategy can be used to fabricate the remarkably nonwetting system on various substrate.

Acknowledgements Authors gratefully acknowledge Universitas Andalas for financial support (contract number: T/58/UN.16.17/PP.IS. KRP2GB/ILPPM/2019).

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

Publisher’s note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

References

27. Xu B, Cai Z (2008) Fabrication of a superhydrophobic ZnO nanorod array film on cotton fabrics via a wet chemical route and