



Preparation of robust anti-smudge coatings via electrophoretic deposition



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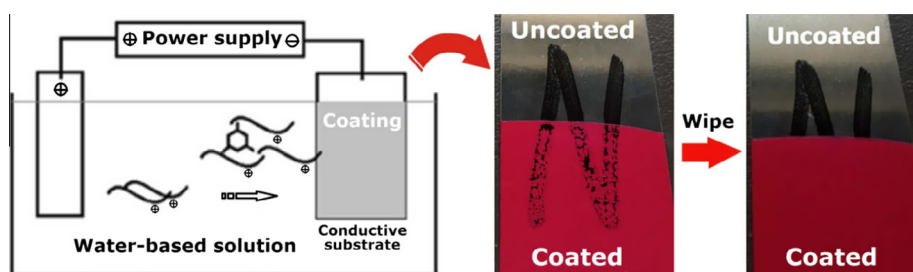
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HIGHLIGHTS

- A robust anti-smudge coating was fabricated using a water soluble precursor.
- EPD was demonstrated as a new strategy for preparing anti-smudge coatings.
- The coating provides substrates with excellent protection.

GRAPHICAL ABSTRACT



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ABSTRACT

Anti-smudge (oil- and water-repellent) coatings that incorporate low surface energy components are typically prepared from solvent-based precursors. Herein, we demonstrate for the first time, a robust and smooth anti-smudge coating that can be fabricated using a water-soluble precursor. Furthermore, the solubility of the novel precursor provides this aqueous electrophoretic deposition strategy with potential as a new strategy for the preparation of anti-smudge coatings for conductive substrates. The coating has outstanding durability, it provides substrates with excellent protection against exposure to harsh conditions, and it can be prepared in various colors. This newly developed strategy can be readily applied onto a diverse range of conductive substrates.

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1. Introduction

Many natural organisms rely on amphiphobic skins to remain clean and free of debris. These natural surfaces have inspired researchers to design and fabricate various artificial liquid repellent materials [1,2]. anti-smudge coatings are highly repellent against water- and oil-borne contaminants, due to their low surface energies [3,4]. The widespread application of these coatings may allow us to have a cleaner world, and to save a lot of time

and money that would otherwise be devoted toward cleaning. The anti-smudge properties of the coatings can be achieved by the use of a rough surface [5–7] (particularly one bearing both micro- and nanoscale roughness as well as re-entrant structures) or (to a lesser degree) by the use of a smooth surface [8,9]. On rough surfaces, the static contact angles of water and oil droplets can exceed 150° (exhibiting superamphiphobicity), and thus it is difficult for liquids to spread on or wet these surfaces [10–12]. Although many attempts have been made to improve these superamphiphobic coatings, there are still unresolved challenges that need to be addressed [13–16], such as their limited durability (the intricate textures of these coatings are prone to damage

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[17,18] and they exhibit poor adhesion to the substrates [19]) and their optical clarity (due to light scattering that occurs as a result of their roughness) [20,21]. On the other hand, by avoiding the use of rough surfaces required for extremely high contact angles, a compromise can be reached through the use of smooth surfaces to yield anti-smudge surfaces exhibiting moderate contact angles while still remaining clean and free of debris after exposure to test liquids [3]. In addition, these smooth anti-smudge coatings are not plagued by the challenges that are typically encountered with rough surfaces, such as poor durability and transparency.

Currently, anti-smudge coatings typically require tailor-made fluorinated compounds (which are insoluble in water) [3–8] and utilize costly solvents such as α,α,α -trifluorotoluene [8] and tetrahydrofuran [9]. To the best of our knowledge, there have been very few liquid repellent rough surfaces [22] and no robust smooth anti-smudge coatings prepared from water-soluble precursors, although a water-based component is highly desirable in order to reduce the cost and environmental footprints of these materials. In addition, the availability of a water-soluble precursor is a key requirement for the industrial aqueous electrophoretic deposition (EPD) strategy to become applicable for the preparation of anti-smudge coatings.

The materials that can be coated via industrial aqueous EPD techniques are limited to conductive substrates such as various alloys, while this processing method has recently gained increasing interest in the industrial because of its key advantages [23]. For example, EPD is significantly more environmentally-friendly and cost-effective than existing techniques, while also being safer and less labor-intensive than many other coating strategies [24]. Furthermore, EPD techniques can be used to fabricate a coating with a very uniform thickness without the formation of pores either within the cavities or on the external surfaces of the substrate, thus it has the potential for the production of anti-smudge coatings for conductive substrates with complex shapes [25,26]. It is noteworthy that conductive alloys with complex shapes are widely used in kitchens, as automotive components, as well as in industrial machines, and they are prone to contamination.

Although the presence of hydrophilic moieties can inhibit anti-smudge properties and the use of water as a solvent is known to weaken the adhesion between a coating and its substrate due to dewetting effects during drying [22,27], we have recently discovered that an anti-smudge coating with outstanding adherence and endurance can be achieved from a water-soluble precursor with a carefully designed structure. We report herein a novel and highly practical water-soluble polyacrylate (WPA) that is applicable for EPD, and demonstrate the superior properties of the resultant anti-smudge coatings that can be applied onto conductive substrates. The WPA had a complex structure, and the monomers involved were selected for various reasons (Fig. 1a). For example, the poly(methyl methacrylate) (PMMA) components were incorporated to enhance the coating transparency, since PMMA is a transparent polymer that is widely used in optical devices [28,29]. Meanwhile, butyl acrylate (BA) was selected for its low glass transition temperature (T_g), and thus the thermal properties of the resultant polyacrylate could be adjusted [30]. 2-(Dimethylamino) ethyl methacrylate (DMAEMA) was selected to provide water-solubility, which was necessary for the EPD protocol. Meanwhile, 2-hydroxypropyl methacrylate (HPMA) units were chosen for their hydroxyl groups that enabled them to react with the polyisocyanate component (blocked hexamethylene diisocyanate trimer, BHDIT, Fig. 1b) in order to prepare the desired heavily crosslinked coating matrix. Styrene was used as a monomer to ensure that WPA was compatible with various aromatic colorants, and to thus accommodate the fabrication of colorful coatings [31,32]. Additionally, 2-(perfluorohexyl)ethyl methacrylate (FHMA) and tri(isopropoxy)vinylsilane (TIVS) were selected as environmentally-

friendly components with low surface tensions [33,34]. Furthermore, due to a potential thermodynamic penalty, the surface bearing CF_3 groups generated from FHMA will not undergo reconstruction during contact with the liquids [35]. TIVS is an inexpensive and readily available commodity coupling agent that enables the coating to bind strongly to metal substrates [36].

2. Experimental

2.1. Materials

Methyl methacrylate (MMA), butyl acrylate (BA), 2-(dimethylamino)ethyl methacrylate (DMAEMA), 2-hydroxypropyl methacrylate (HPMA), styrene, 2,2'-azobis(2-methylpropionitrile) (AIBN), and methyl ethyl ketoxime (MEKO) were purchased from Hersbit Chemical Co. Ltd., and were of analytical reagent (A.R.) grade. Hexamethylene diisocyanate trimer (HDIT, NCO content of 21.8 ± 0.3 wt%) was purchased from Wanhua Chemicals Co., Ltd. and was used as received. 2-(Perfluorohexyl)ethyl methacrylate (FHMA, >99%) was purchased from Hengtong Fluorine Co., Ltd. and purified via vacuum distillation prior to use. Tri(isopropoxy)vinylsilane (TIVS, >99%) was kindly supplied by Ark (FoGang) Chemicals Industry Co., Ltd. and distilled under reduced pressure prior to use. 2-Hydroxypropanoic acid, propylene glycol monomethyl ether (PGME), propylene glycol 1-monomethyl ether 2-acetate (PGMEA), diiodomethane, hexadecane, and rhodamine were all of A.R. grade and were purchased from Tianjin Damao Chemical Reagent Factory. Sandpaper (Standard glasspaper, Grit No. 400) was purchased from a local store.

2.2. Synthesis of the WPA

WPA was synthesized via free radical polymerization with the use of various monomers (Fig. 1a). In a typical procedure, a calculated amount of PGME (20.0 g), PGMEA (40.0 g), and AIBN (0.4 g) were added into a 250 mL four-necked round-bottom flask equipped with feeding inlets, a mechanical stirrer, a thermometer, and a reflux condenser. The reaction mixture was purged under a flow of nitrogen for 30 min at room temperature, and the flask was subsequently heated to 90 °C. The mixture of MMA (20 g, 0.20 mol), BA (30 g, 0.23 mol), DMAEMA (10 g, 0.064 mol), HPMA (12 g, 0.083 mol), styrene (12 g, 0.12 mol), and AIBN (0.40 g) was added dropwise into the flask over a period of ~2 h. The mixture of FHMA (8.0 g, 0.019 mol) and TIVS (8.0 g, 0.034 mol) was then added dropwise into the above flask over a period of ~0.5 h. AIBN (0.40 g) was subsequently added into the above mixture, and the reaction was continued for an additional 12 h at 90 °C. This reaction mixture was finally cooled to room temperature, and the DMAEMA units were neutralized using 2-hydroxypropanoic acid.

2.3. Synthesis of the BHDIT

A calculated amount of HDIT (50 g, 0.099 mol) was added under a nitrogen flow into a flask equipped with feeding inlets, a mechanical stirrer, a thermometer, and a reflux condenser connected to a drying tube. MEKO (26 g, 0.030 mol) was added dropwise into the above flask under vigorous stirring. The mixture was left to react overnight at room temperature to yield BHDIT.

2.4. Preparation of the coatings

The procedure employed to prepare the coatings via the EPD strategy is illustrated in Fig. 1c. WPA (9.0 wt%), BHDIT (2.3 wt%), and distilled water were mixed together. The aluminum and tin plates were subjected to EPD by submerging them into this aque-

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