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Formation, structure, and reactivity of amino-terminated organic films on silicon substrates

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ABSTRACT

Amino-functionalized organic films were prepared by self-assembling 3-aminopropyltriethoxysilane (APTES) on silicon wafers in either anhydrous toluene or phosphate-buffered saline (PBS) for varied deposition times. Fourier transform infrared spectroscopy (FTIR) and ellipsometry have shown that the structure and thickness of APTES films are governed by the deposition time and reaction solution. Deposition from an anhydrous toluene solution produces APTES films ranging from 10 to 144 Å in thickness, depending on the reaction time. FTIR spectra indicate that film growth initially proceeds by adsorption of APTES to the silicon surface followed by siloxane condensation, and after an extended period of time APTES molecules accumulate on the underlying APTES film by either covalent or noncovalent interactions. In contrast, spectroscopically indistinguishable APTES films in thickness ranging from 8 to 13 Å were formed when deposition was conducted in aqueous solutions. Measured water contact angles indicate that APTES films deposited in aqueous solutions are more hydrophilic compared to those prepared in toluene solutions. Fluorescence measurements revealed that APTES films prepared in toluene solutions contain more reactive surface amino groups by ca. 3 to 10 times than those prepared in aqueous solutions for the identical reaction time.

1. Introduction

3-Aminopropyltriethoxysilane (APTES) is one of the most frequently used organosilane agents for the preparation of amino-terminated films on silicon substrates [1–13]. A general consensus regarding the APTES film formation is that silanization begins with the hydrolysis of ethoxy groups in APTES, a process catalyzed by water, leading to the formation of silanols. APTES silanols then condense with surface silanols forming a monolayer of APTES via a lateral siloxane network in which amino groups are oriented away from the underlying silicon surface. Many experimental results, however, have suggested that this is an oversimplified description of the idealized reaction and that the actual process is far more complex and sensitive to reaction conditions [8,13–23].

The complexity of the APTES silanization reaction mainly stems from the presence of a reactive amino group in APTES and its inherent propensity to enter into competing reactions. The amino groups in adsorbed APTES interact with silanols present on the silicon surface and/or in the adjacent hydrolyzed APTES via hydrogen bonding or electrostatic interactions [14,18,19,24,25]. This considerably reduces the number of available silanol groups on both the silicon surface and the APTES for further siloxane condensation. In addition, polymerization of APTES gives rise to the formation of multiple APTES layers whose structure and reactivity are likely different from those of an APTES monolayer. These are significant issues when preparing APTES films since the availability and reactivity of surface amino groups are crucial to many applications including the construction of biomedical devices [1–5,9,11,23,26].

APTES films on silicon substrates have been characterized by numerous analytical techniques including X-ray photoelectron spectroscopy (XPS) [17,18,27–30], Fourier transform infrared spectroscopy (FTIR) [31–39], ellipsometry [17,22,40], scanning probe microscopy [8,17,28,29], and contact angle measurements [18,30,41,42]. In particular, Fourier transform infrared spectroscopy with various sampling techniques has been popularly used to extract structural information regarding organic films of various thicknesses on silicon wafers.

Most conventional FTIR data acquisition techniques include transmission [31,32], external reflection [33–37], and internal reflection [38,39]. Both transmission and external reflection methods generally produce infrared spectra with a low signal-to-noise ratio. Hence, these methods require a large number of scans (∼500–1500) and sensitive detection systems (e.g., MCT) to produce infrared spectra of good quality. Although the traditional internal reflection method has provided a feasible way to obtain infrared data of organic films with higher signal-to-noise ratios, it requires direct deposition of organic films on infrared-transferable crystals.

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Compared to these conventional sampling methods, an exceptional enhancement of absorbance signals in FTIR spectra is achieved when organic layers on silicon substrates are brought into direct contact with high refractive index infrared-transferable crystals (e.g., germanium). This improved sampling technique, a grazing-angle attenuated total reflection (GATR) method, has successfully produced FTIR spectra of monolayers of organic thin films on silicon substrates with enhanced sensitivity [43–46].

In this work, we investigated the effect of the reaction solution and deposition time on the structure and reactivity of APTES films via FTIR-GATR, ellipsometry, contact angle measurement, and fluorescence microscopy. APTES films were produced in two different solutions, anhydrous toluene and phosphate-buffered saline (PBS), for varied deposition times. Based on experimental data presented herein, the structure and reactivity of APTES films on silicon substrates are described, the significance of solvent and reaction time is discussed, and finally mechanisms of APTES film formation in two solutions are compared in this paper.

2. Experimental

Deionized water used for the preparation of chemical solutions and in the cleaning of the experimental apparatus was produced by a Millipore water purification system with a minimal resistivity of 18.0 MΩ cm. 3-Aminopropyltriethoxysilane (APTES, 99.0%, Acros), 1,1,1,3,3,3-hexamethyldisilazane (HMDS, 98.0%, Acros), and anhydrous toluene (99.8%, extra dray, water < 30 ppm, Acros) were purchased and used as received without further purification.

P-doped, (100)-oriented silicon wafers (prime grade, 7–21 Ω cm resistivity, ∼0.5 mm thickness, 5-inch diameter) were cut into ca. $10 \times 10\ \text{mm}^2$ squares for FTIR and fluorescence measurements and ca. $25 \times 25\ \text{mm}^2$ squares for ellipsometric and contact angle measurements, respectively. Silicon substrates were sonicated in acetone for 10 min, a mixture of acetone (50%) and ethanol (50%) for 10 min, and rinsed with a copious amount of deionized water. After that, silicon wafers were cleaned in freshly prepared Piranha solution (a mixture of 30% H_2O_2 and 70% of H_2SO_4) for at least 5 h, rinsed exhaustively with deionized water, and dried in a stream of nitrogen gas before APTES deposition.

Two types of solvents, anhydrous toluene (water < 30 ppm) and phosphate-buffered saline (PBS, pH 7.2 ± 0.1, and ionic strength 10 mM), were used to prepare APTES solutions with a concentration of 2.0% (v/v). APTES films were prepared by incubating clean silicon wafers in APTES solutions for 15 min, 1, 4, or 24 h. For a given deposition time, silicon wafers for FTIR, ellipsometry, contact angle measurements, and fluorescence microscopy were incubated simultaneously in the same reaction vessel. All glass reaction vessels were passivated twice by the use of vaporized HMDS at 100 °C before use. After the controlled deposition, silicon wafers were sonicated twice in APTES-free solvents for 10 min to remove loosely physisorbed APTES. Then, these wafers were dried by the use of a stream of nitrogen gas before data collection.

Ellipsometric thickness measurements were conducted with a Gaertner L116A automatic ellipsometer equipped with a HeNe laser (632.8 nm). Optical constants of $n = 3.865$ and $k = 0.020$ for Si, $n = 1.465$ and $k = 0$ for both organic layers and the underlying SiO_2 layer were used [8,47]. The thickness of the silicon oxide layer was measured before APTES film deposition and subtracted from the total layer thickness after the formation of APTES layers as described previously [8,47,48]. For each preparation condition, at least 10 spots from three different silicon wafers were measured. Three standard samples having an oxide layer thickness of 22, 100, and 500 Å were used to confirm the proper workability of our ellipsometer before data collection.

Although details of experimental setups for FTIR-GATR vary, optical configurations similar to our system are described elsewhere [43,44,46]. Briefly, a VeeMax II sampling stage (Pike Technologies, Madison, WI) equipped with a 60° germanium (Ge) ATR crystal and a high-pressure clamp was placed in the sample compartment of a Nicolet Magna 550 FTIR spectrometer. A high-pressure swivel clamp (diameter ∼ 7.8 mm) was used to apply even and constant pressure (∼35 lbs/cm^2) to the sample during FTIR data acquisition. The angle of incident infrared was set ca. 60° with respect to the surface normal of Ge crystal. Silicon substrates with APTES film were placed (face down) between the Ge crystal and the tip of the high-pressure clamp. Each FTIR spectrum represents the average of 200 scans at 4 cm^{-1} resolution. A *p*-polarized infrared beam was used and the output signal was collected with a deuterated triglycine sulfate (DTGS) detector.

Water contact angle was measured via the static sessile drop method using a manual goniometer (Ramé-Hart Instrument Co., Model 50-00) as described previously [30,41,42]. Measurements were made 10 s after dropping 10 µl of deionized water using a microsyringe. For each sample, at least 10 measurements were used to obtain averages and standard deviations reported in this paper.

The reactivity of APTES films was estimated by fluorescence measurements after carboxyfluorescein ($\lambda_{\text{excitation}} = 494$ nm and $\lambda_{\text{emission}} = 521$ nm) was conjugated to surface amino groups. The conjugation reaction was carried out using 5(6)-carboxyfluorescein, *N*-hydroxysuccinimide ester (Marker Gene Technologies, Inc., Eugene, OR) at a concentration of 100 µg/mL in a mixture of 0.1 M sodium bicarbonate buffer (pH 8.2) and DMSO (5.0%, v/v) for 3 h. After reactions, silicon wafers with APTES films and conjugated dyes were sonicated twice in deionized water for 10 min and dried with N_2 gas prior to fluorescence measurements. Fluorescent measurements were conducted using an Olympus BX51 fluorescence microscope and images were recorded with a digital color CCD camera (Diagnostic Instruments Model 1.3.0). Recorded images were converted into digitalized intensities, subtracted from a background measurement from an APTES film without dye conjugation, and finally produced averages and standard deviations of fluorescence intensities for each APTES film.

3. Results

In a first set of experiments, the thickness of APTES films prepared for varied deposition times was measured and the results are listed in Table 1. Ellipsometric data indicate that the thickness of APTES films depends on the reaction time and solution. In both toluene and PBS solutions, thicker APTES films were prepared when deposition was conducted for longer periods of time. While the thickness of APTES films prepared in toluene ranged from 10 to 144 Å, those from PBS were much thinner (8–13 Å). An APTES film roughly equivalent in thickness to a monolayer (∼7–9 Å) was prepared in PBS when reaction time was limited (15 min and 1 h), while multilayers of APTES films were produced in toluene [25,40].

Infrared data of APTES films were collected after sonication. For APTES films produced in toluene solutions, all FTIR spectra show similar features between 2800 and 3000 cm^{-1} which originate from several CH stretch modes of the APTES backbone and ethoxy

Table 1
The layer thickness (Å) of APTES films prepared in toluene and PBS for 15 min, 1 h, 4 h, or 24 h followed by sonication two times in APTES-free reaction solutions for 10 min.

Reaction time	Toluene	PBS
15 min	10 (1)	8 (1)
1 h	17 (1)	9 (1)
4 h	61 (2)	10 (1)
24 h	144 (3)	13 (1)

Standard deviations are in the parentheses.

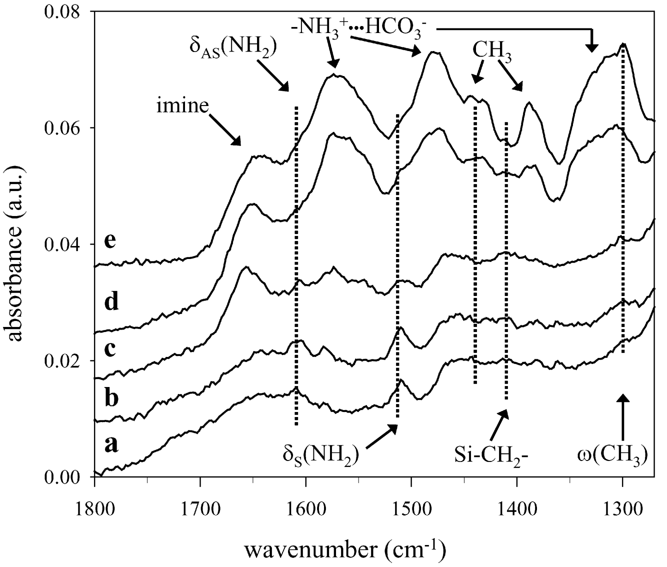

Fig. 1. FTIR spectra of APTES films produced in anhydrous toluene solutions with a deposition time of 15 min (a), 1 h (b), 4 h (c), and 24 h (d) followed by sonication twice in toluene for 10 min. The measured thickness of these films was 10, 17, 61, and 144 Å. For comparison, a FTIR spectrum of an APTES film with a thickness ∼1300 Å prepared by spinning-cast is also included (e).

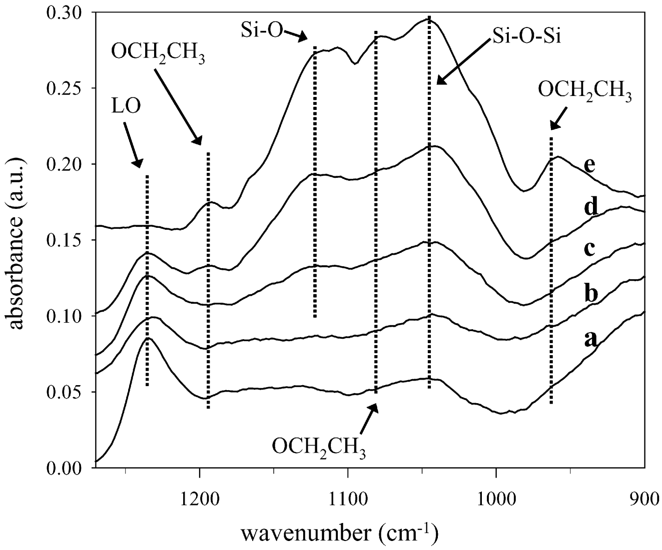

Fig. 2. FTIR spectra from the same APTES films shown in Fig. 1. Deposition times were 15 min (a), 1 h (b), 4 h (c), and 24 h (d). An FTIR spectrum of an APTES film with a thickness ∼1300 Å was included for comparison (e).

groups (FTIR spectra are available in the Supporting information). Although toluene can contribute signal intensities in this region as well as around 730 and 695 cm^{-1}, none of these peaks were observed in any of the FTIR spectra presented in the paper. This fact indicates that the amount of entrapped toluene in APTES films is too small to be detected by our FTIR studies. Around 3250 and 3350 cm^{-1}, the symmetric and asymmetric NH stretch modes from amino groups in APTES films are very weak in intensity regardless of the film thickness [14,49].

The most important structural information regarding APTES films is found between 1800 and 900 cm^{-1}. FTIR spectra in the range of 1800 to 1270 cm^{-1} are shown in Fig. 1. An FTIR spectrum of an APTES film prepared by spinning-casting on a silicon wafer (thickness ∼ 1300 Å) is included for comparative purpose (see Fig. 1e). A vibrational mode around 1655 cm^{-1} is due to the presence of an imine group formed by the oxidation of an amine bicarbonate salt and its intensity is closely related to the film thickness [16,17,20,21]. There are several vibrational modes with various absorptions between 1610 and 1460 cm^{-1} mainly originating from NH_2 groups in the APTES film. Two weak vibrational modes around 1610 and 1510 cm^{-1} are observed for films with the thickness less than 61 Å (Fig. 1a, 1b, and 1c), but they are not clearly observed in spectra of thicker APTES films (Fig. 1d and 1e). These modes are assigned to the asymmetric and symmetric deformation modes of NH_2 groups in adsorbed APTES located near the silicon surface [19–21]. These NH_2 groups are known to associate with silanols present on the silicon surface. A proton transfer from acidic surface silanols to adjacent basic NH_2 groups may give rise to of the formation of $SiO^-\ldots H\ldots NH_2^+$ species as was proposed previously [19,25].

Two dominating vibrational modes are found around 1575 and 1485 cm^{-1} in Fig. 1. It is noted that the intensity of the modes at 1575 and 1485 cm^{-1} increases as the film thickness increases. However, the intensity of these two modes is comparable for the two films with thicknesses of 144 and 1300 Å (see Fig. 1d and 1e). This observation indicates that a major contribution of these vibrational modes at 1575 and 1485 cm^{-1} arises from surface amino groups, rather than those present in the bulk film. It was also observed that the intensity of these modes was attenuated when film deposition was conducted in a N_2 atmosphere [15,16,49]. Therefore, the vibrational modes at 1575 and 1485 cm^{-1} arise when surface amino groups form bicarbonate salts in a reaction with atmospheric CO_2 as was reported previously [14,21].

In Fig. 1, c, d, and e, the asymmetric and symmetric deformation modes of the CH_3 group from ethoxy moieties of APTES are observed around 1440 and 1390 cm^{-1}, respectively [14,21]. The presence of these two modes indicates the existence of ethoxy groups in adsorbed APTES, presumably due to incomplete siloxane condensation or the presence of physically adsorbed unhydrolyzed APTES. The bending mode of the methylene group adjacent to Si in APTES ($-Si-CH_2-$) is found around 1410 cm^{-1} [14,20,21]. The mode near 1330 cm^{-1} is due to the presence of an amine bicarbonate salt [15,16,49]. A band at 1300 cm^{-1} is assigned to the CH_2 wagging mode from the APTES backbone [14,21]. Vibrational modes at 1440, 1410, 1390, and 1330 cm^{-1} are not clearly observable in Fig. 1, a and b, indicating that all ethoxy groups in APTES films close to the silicon surface are hydrolyzed, allowing for siloxane condensation between surface silanols and neighboring surface-bound APTES.

FTIR spectra in the range from 1270 to 900 cm^{-1} for APTES films prepared from toluene solutions are presented in Fig. 2. The mode around 1235 cm^{-1} is assigned to the longitudinal optical (LO) mode which is generally observed from a thin reflective silicon oxide film (5–100 Å) [43,44,50]. The intensity of this LO mode decreases as the APTES layer thickness increases. This LO mode was barely observable when the film thickness was about ∼1300 Å (Fig. 1e). Vibrational modes around 1195, 1080, and 960 cm^{-1} arise from unhydrolyzed ethoxy moieties in APTES ($-OCH_2CH_3$) [14,20,21]. The presence of these modes, as well as those modes around 1440 and 1390 cm^{-1} in Fig. 2d and 2e, indicates incomplete siloxane condensation of adsorbed APTES in films grown for extended periods of time (e.g., 24 h). The peak located around 1125 cm^{-1} is attributed to Si–O moieties of either polymerized or physisorbed APTES. Note that the intensity of this mode is proportional to the APTES layer thickness and increases from a to d in Fig. 2. Compared to the previously reported FTIR spectra, its position depends on several factors including the presence of ethoxy groups [14,17,20,21]. The strong mode around 1045 cm^{-1} is attributed to siloxane groups (Si–O–Si) from (i) the silicon substrate, (ii) condensed APTES with the silicon surface, and (iii) polymerized APTES [14,17,20]. This mode is also proportional to the APTES layer thickness and increases from a to d in Fig. 2.

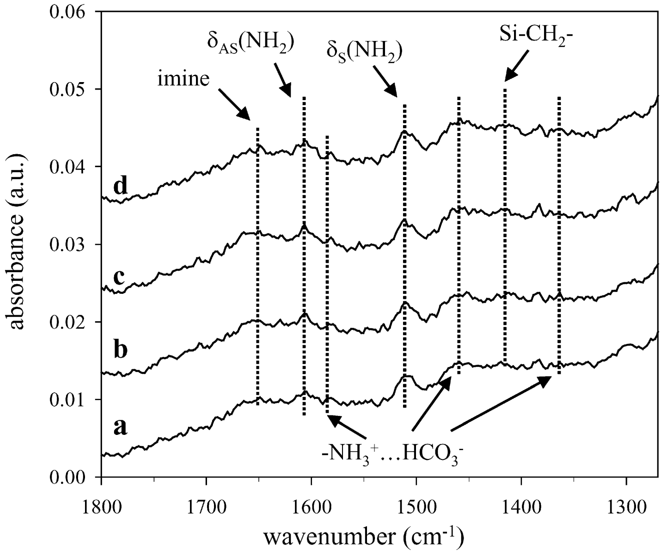

Fig. 3. FTIR spectra of APTES films produced in PBS with the deposition time of 15 min (a), 1 h (b), 4 h (c), and 24 h (d) following a 20 min sonication in PBS. Measured thicknesses of these films were about 8, 9, 10, and 13 Å, respectively.

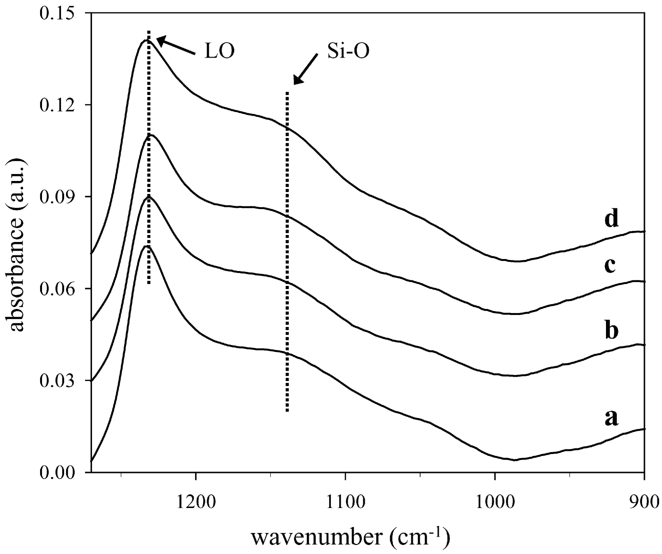

Fig. 4. FTIR spectra from same APTES films shown in Fig. 3, but in the range 1270 to 900 cm^{-1}.

All FTIR spectra of the APTES films produced in PBS contain CH stretch modes between 2800 and 3000 cm^{-1} similar to those observed from spectra of APTES films prepared in toluene solutions. One noticeable difference is the existence of a broad peak around 3300 cm^{-1} presumably due to adsorbed water molecules on APTES films (see the Supporting information). FTIR data of these APTES films in the region of 1800 to 1270 cm^{-1} are shown in Fig. 3. Compared to spectra from APTES films prepared in toluene solutions (Fig. 1), all spectra contain similar vibrational features, intensities, and positions regardless of the reaction time. The presence of the mode at 1510 cm^{-1} along with the absence of observable peaks around 1575 and 1485 cm^{-1} suggests that virtually all amino groups in APTES films are associated with surface silanols. Fig. 4 shows FTIR data in the region of 1270 to 900 cm^{-1} from the same APTES films. Again, all spectra are similar to each other with respect to the LO mode and weak shoulders around 1235 and 1125 cm^{-1}. These FTIR spectra indicate that APTES films are structurally comparable to each other regardless of the reaction time when adsorption was conducted in aqueous solutions.

Table 2
Measured water contact angles (°) for APTES films prepared in toluene and PBS for varied deposition times.

Reaction time	Toluene	PBS
15 min	50.7 (1.6)	37.9 (2.1)
1 h	52.4 (1.8)	38.2 (1.8)
4 h	53.0 (1.9)	41.1 (1.8)
24 h	54.7 (1.6)	44.2 (0.9)

Standard deviations are in the parentheses.

Table 3
Fluorescence intensities from carboxyfluorescein-conjugated APTES films prepared in toluene and PBS for varied deposition times.

Reaction time	Toluene	PBS
15 min	17.7 (1.7)	5.4 (0.9)
1 h	32.0 (4.0)	6.5 (0.6)
4 h	74.3 (9.5)	6.8 (1.1)
24 h	90.1 (8.5)	8.9 (1.0)

Standard deviations are in the parentheses.

Table 2 shows static water contact angles measured on APTES films prepared for varied deposition times from two different reaction solutions. Contact angle data indicate that the wettability of APTES primarily depends on the thickness of APTES films. The surface becomes more hydrophobic as the APTES film thickness increases. For APTES films prepared in toluene solutions, water contact angles range from 51° to 55° depending on the film thickness. However, contact angles from APTES films deposited in aqueous solutions are between 38° and 44°.

In a final set of experiments, the reactivity of amino groups on APTES films was estimated by fluorescence measurements. Table 3 shows fluorescence intensities of carboxyfluorescein molecules conjugated to APTES films produced in either toluene or aqueous solutions for varied deposition times as described above (fluorescence images are provided in the Supporting information). Fluorescence intensities of APTES films prepared in toluene were greater than those prepared in PBS for the same deposition time by ca. 3 to 10 times.

4. Discussion

From ellipsometric measurements and FTIR spectra, it was found that APTES films on silicon substrates adopt different structures depending on the reaction solution and deposition time. Such structural differences are attributed to the difference in the APTES adsorption process and film formation mechanism as reported previously [9]. In a toluene solution, ethoxy groups in APTES are not completely hydrolyzed. We suppose that hydrogen bonding between the amino group in APTES and surface silanol plays an important role in initial APTES adsorption as indicated by the presence of vibrational features at 1610 and 1510 cm^{-1} in FTIR spectra. Once near the surface, a fraction of those ethoxy groups in adsorbed APTES molecules are hydrolyzed by trace amounts of surface-bound water and subsequently condense with surface silanols and other adjacent surface-bound APTES giving rise to flat lateral film growth as was proposed previously (see Fig. 5a) [9].

Unreacted ethoxy (or hydrolyzed silanol) groups of adsorbed APTES may lead to condensation with dissolved APTES giving rise to vertical film growth. However, the condensation is slower due to the absence of water. As the reaction time increases, more APTES accumulates via covalent condensation and/or physisorption, leading to thicker films (see Fig. 5b). Previous studies by atomic force microscopy (AFM) and scanning electron microscopy (SEM) have observed that thinner APTES films are generally flat and homogeneous, but aggregates are found on thicker APTES films produced for extended reaction times [8,17,28,29]. These thicker

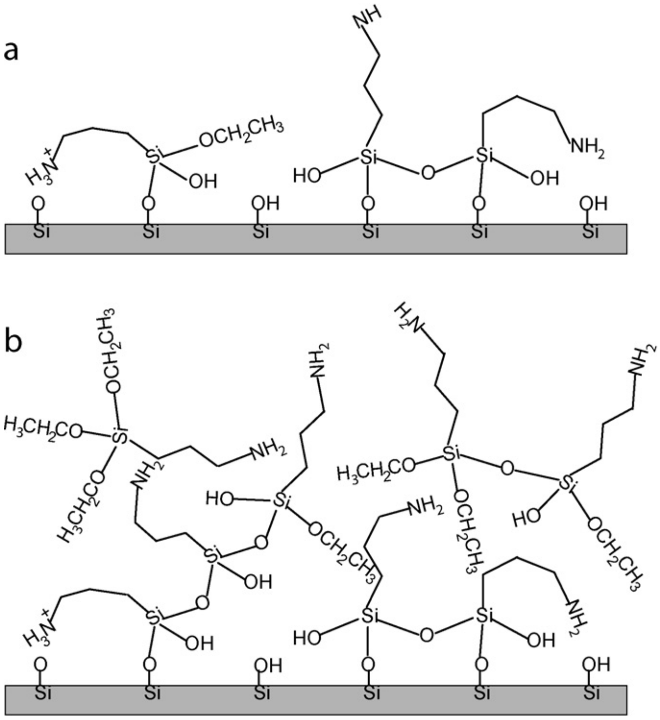

Fig. 5. Schematic representation of adsorbed APTES on a silicon wafer in an anhydrous toluene solution. Ethoxy groups of adsorbed APTES are eventually hydrolyzed and form siloxane bonds with surface silanols (a). Further siloxane condensation between surface bound APTES and dissolved APTES and/or noncovalent adsorption of dissolved APTES results in the formation of APTES multilayers on the silicon wafer (b).

APTES films contain a mixture of condensed APTES and aggregates of physisorbed APTES. Although a major portion of physisorbed APTES was removed by sonication, we believe that a significant amount of partially condensed and/or noncondensed APTES remains in the film.

In an aqueous solution, most ethoxy groups in APTES are readily hydrolyzed to silanols. Since the silicon oxide surface ($\mathrm{p}K_a < 5$) is negatively charged and the amino group in APTES is partially protonated in aqueous solutions at pH ~7.2, APTES likely adsorbs to the silicon surface via electrostatic interactions [51]. We suppose that a majority of adsorbed APTES adopts a conformation with its amino group directing toward a negatively charged silicon surface while negative (or neutral) silanols point away from the silicon surface as shown in Fig. 6a. This is supported by our contact angle experiments and fluorescence measurements shown in Tables 2 and 3. The measured contact angle from an APTES film produced in toluene solutions for 15 min, 50.7°, is greater than that from an APTES film produced in aqueous solutions for 4 h, 41.1°, despite comparable thicknesses of the two APTES films, ~10 Å. The presence of charged APTES and/or APTES with silanol groups directing away from the surface is likely responsible for the formation of the more hydrophilic APTES films which were produced by deposition in aqueous solutions.

Fluorescence intensity from the APTES film prepared in toluene for 15 min (thickness 10 Å) is greater by about two and half times than that from the APTES film of 10 Å prepared in an aqueous solution for 4 h. This suggests that less surface amino groups are available on thicker APTES films prepared in aqueous solutions presumably due to different orientations of amino groups on silicon surfaces.

It seems that siloxane condensation between silanol groups of adsorbed APTES and surface silanols is not as feasible in aqueous solutions. Instead, further deposition of charged APTES in an aqueous solution is expected via electrostatic interactions and/or hydrogen bonding, forming multilayers as described in Fig. 6b. Unlike those APTES films prepared in toluene, APTES layers formed in water consist of weakly bound APTES via electrostatic interactions/or hydrogen bonding. This may explain our observation that APTES films prepared in aqueous solutions are thinner than those prepared from toluene solutions at equal reaction times after sonication.

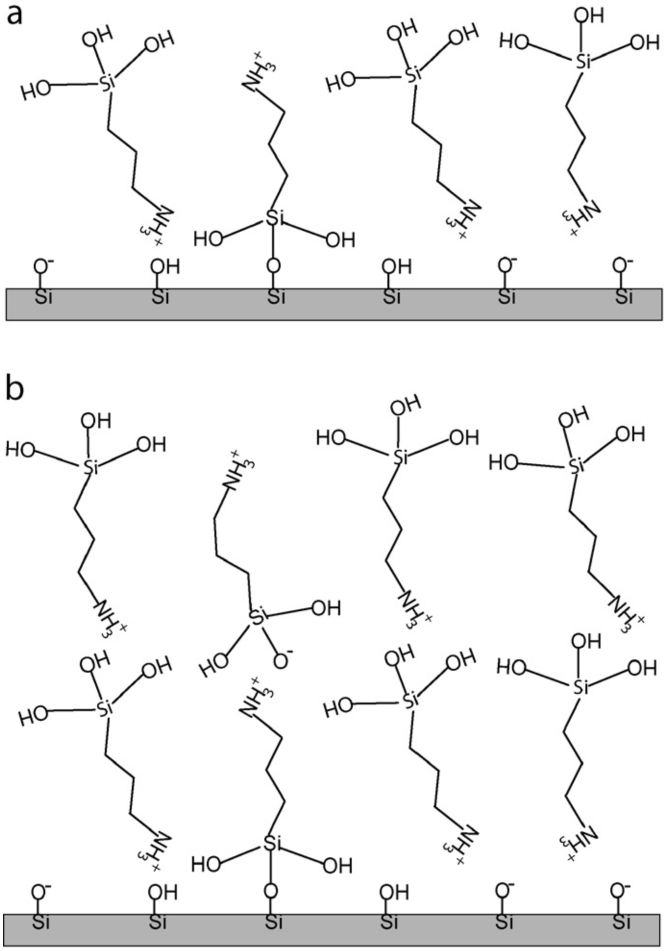

Fig. 6. Schematic representation of adsorbed APTES on a silicon wafer in PBS. Initial APTES adsorption is driven via electrostatic interactions between positively charged amino groups and the negatively charges silicon surface in PBS with pH ~7.2. Some of the adsorbed APTES condenses with surface silanols forming siloxane linkages. Further deposition is also expected to proceed by electrostatic interactions and/or hydrogen bonding to form multilayers of APTES (b).

5. Summary and conclusions

Ellipsometry, FTIR-GATR, contact angle measurements, and fluorescence microscopy show that the structure, thickness, wettability, and reactivity of APTES films are affected by preparation conditions such as the deposition time and the choice of reaction solutions due to differences in the APTES adsorption process and film growth mechanisms. In an anhydrous toluene solution, APTES films grow by both covalent and noncovalent adsorption of APTES. There exist at least three different types of APTES in these films; APTES directly condensed on the silicon surface, APTES condensed with other APTES, and physisorbed APTES. However, APTES films grow by electrostatic interactions/or hydrogen bonding when deposited from aqueous solutions. These films are not stable and are easily disrupted by sonication, leaving one or two monolayers of APTES on the silicon surface which are more hydrophilic than multilayers of APTES formed in anhydrous toluene solutions. Additionally a major portion of the surface amino groups from either condensed or physisorbed APTES in multilayers readily forms bicarbonate salts in a reaction with CO_2 in the air. The reactivity of these APTES films evaluated by fluorescence measurements shows that APTES films prepared in toluene solutions contain more surface reactive amino groups.

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Supporting information

Supplementary data for this article may be found in the online version at DOI: 10.1016/j.jcis.2008.09.031. The data include two FTIR spectra in the range between 3800 and 2600 cm^{-1} from APTES films from both toluene and aqueous solutions with different deposition times, three FTIR spectra of toluene at the silicon/germanium interface, and eight fluorescence images from APTES films from both toluene and aqueous solutions with different deposition times.

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