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The effect of organo-functional silanes on the adhesion of epoxide resins to ITO glass

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Abstract—In this study we investigated five different types of organo-functional silanes containing vinyl, epoxy, amino, methacrylic and acrylic groups in the cationic polymerization of epoxide resin and studied their effects on epoxide adhesion strength to indium tin oxide (ITO) substrate *via* a pull test. The adhesion strength of epoxide resins was found to be in the order vinyl > methacrylic > acrylic > epoxy > amino. The silane containing vinyl group enhanced the adhesion strength, since it was able to promote free-radical polymerization. As to the silane containing amino group, it retarded the photo-polymerization of cationic photoinitiator in the resin and thus the lowest adhesion strength was observed. It was found that the resin sample containing 1 wt% of vinyl group silane possessed the highest adhesion strength of 91.42 kg/cm^2 to ITO glass. An excessive amount of silane added in the resin generated too much H_2O that diluted the concentration of resin and deteriorated the adhesion strength.

Keywords: Adhesion strength; ITO glass; silane functional group; epoxide resins.

1. INTRODUCTION

Due to the advantages, such as low power consumption, high efficiency, wide viewing angle, fast response time, and compact and lightweight nature, organic luminescent devices (OLEDs) are currently one of the promising flat panel displays [1]. At present, indium tin oxide (ITO) is adopted as the anode conductor on glass substrate in OLEDs [2, 3] and, hence, in order to achieve a reliable packaging structure, sufficient adhesion strength of adhesive resins to both glass and ITO is required. Silane coupling agents are commonly adopted as surface modification chemicals and are used primarily as adhesion promoters in composite materials, particularly to achieve strong bonding of polymeric materials to metals, glass and minerals [4]. Typically,

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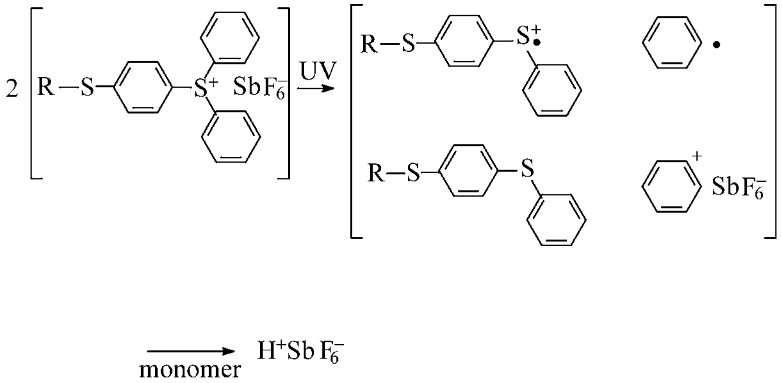
*To whom correspondence should be addressed. Tel.: (886-3) 571-2121, ext. 55338. Fax: (886-3) 572-4727. E-mail:cjjiang@ms.chinmin.edu.tw a silane coupling agent possesses alkoxy groups and an organo-functional group. The alkoxy groups, OR, hydrolyze in the aqueous environment and form covalent bonds with hydroxyl groups on the surface, as well as with other hydrolyzed silane molecules. The organo-functional group, R′, also reacts with the matrix resin [5, 6]. A properly chosen organo-functional group for a specific application increases the interfacial strength significantly. In some cases the adhesion enhancement is due to the formation of covalent linkages with the polymer *via* chemical interactions to produce a so-called interpenetrating polymer network (IPN) [7]. The widespread use of organo-functional silanes prompts an in-depth study of the mechanisms of adhesion enhancement. In this study, the organo-functional group was mainly chosen according to its reactivity and compatibility with the photo-initiator. Typically, silane coupling agents with amine [8], epoxy [9], methacrylic [10], acrylic [11], or vinyl [12, 13] organo-functional groups are used for adhesion enhancement. In this work, these five silanes were added to the base adhesive resin, to form five different epoxide derivatives. Each of these epoxide derivatives was applied onto glass and ITO to form a sandwich specimen for pull test. The results were correlated to different silane additives to explore the effect on adhesion strength.

This work focused on the cationic photo-polymerization reaction of epoxide resin and polyol monomer and the reaction mechanisms are depicted in equations (1)–(6).



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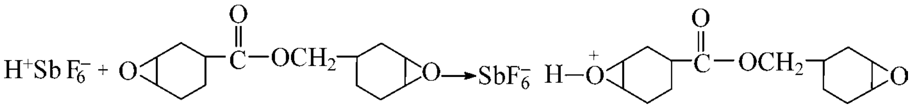

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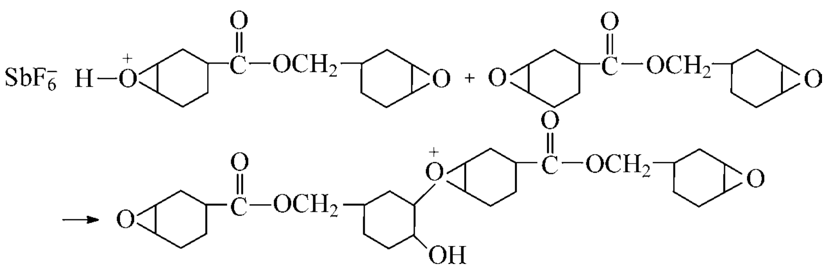

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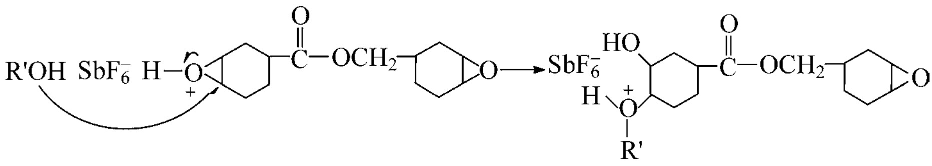
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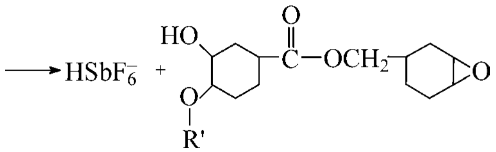
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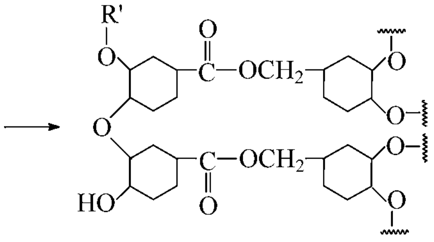
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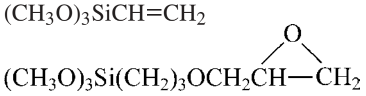
Depending on the method of application, a silane coupling layer normally consists of several monolayers. There are basically four different methods of applying silane coupling agents: deposition from aqueous solutions, deposition from organic solutions, integral blend method, and deposition as a primer [14]. The adhesives for OLED packaging must be solvent-free to prevent outgassing during subsequent curing, we hence adopted the integral blend method to add silanes into the epoxide resin.

2. EXPERIMENTAL

2.1. Materials

3,4-Epoxycyclohexylmethyl 3,4-epoxycyclohexanecarboxylate oligomer, triarylsulfonium hexafluoroantimonate salt photoinitiator and tri(propylene glycol) monomer were purchased from Aldrich. Benzoyl peroxide (BPO) thermal initiator was supplied by Fluka. The silane coupling agents studied were: vinyltrimethoxysilane, 3-glycidoxypropyltrimethoxysilane, 3-aminopropyltrimethoxysilane, 3-acryloxypropyltrimethoxysilane and 3-methacryloxypropyltrimethoxysilane, all supplied by Aldrich. Their chemical structures are shown in Table 1. Each adhesive resin was prepared by mixing dicycloaliphatic epoxide, and the monomer in a 4 : 1 ratio by weight and one of the selected silane coupling agents at 3 wt% for 1.5 h. After that,

Table 1.
Structures and designated names of organo-functional silanes

Organo-functional silane	Chemical structure	Designated name
Vinyltrimethoxysilane	$(CH_3O)_3SiCH{=}CH_2$	vinyl
3-Glycidoxypropyltrimethoxysilane		epoxy
3-Aminopropyltrimethoxysilane	$(CH_3O)_3Si(CH_2)_3NH_2$	amino
3-Acryloxypropyltrimethoxysilane	$(CH_3O)_3Si(CH_2)_3OOCCH{=}CH_2$	acrylic
3-Methacryloxypropyltrimethoxysilane	$(CH_3O)_3Si(CH_2)_3OOCC(CH_3){=}CH_2$	methacrylic

2 g of photoinitiator and 1.5 g of BPO were added into each prepared adhesive resin and stirred for 1.5 h.

The ITO glass with dimensions of 10 mm × 10 mm × 0.7 mm was supplied by Merck Display Technologies Ltd. (Taiwan). The ITO overlayer had a thickness of 200 nm and sheet resistance of 10 Ω/m. The glass block with dimensions of 10 mm × 10 mm × 8 mm was purchased from Liehcher (Taiwan).

2.2. Adhesion test

The glass block was polished into a truncated shape with top surface area equal to 7 mm × 7 mm as shown in Fig. 1a. After applying the adhesive resin on this area, the glass block was glued to the ITO glass to form the pull test specimen. The specimens were cured in a UV oven (CL-1000M which was supplied by UVP, USA) with wavelength ranging from 314 to 340 nm at a power of 40 W for 25 min and then post-cured at 80°C for 1 h to complete the hardening process. Two steel bars were then attached onto the two ends of the sample and set to a pull tester for adhesion strength measurement at the pull rate of 5 mm/min. The pull tester was Model-HT-8116 purchased from Hung Ta Instrument (Taiwan). For each type of adhesive sample, the pull test was repeated five times. Adhesion strengths of adhesive resins were measured by a pull tester as shown in Fig. 1b, which corresponds to the ASTM D897 standard.

2.3. FT-IR and NMR characterizations

The FT-IR samples were prepared by coating silanes and silanes blended with photoiniator on KBr discs and then transferred to a Nicolet Protégé 460 Fourier-transform infrared spectrometer (FT-IR) for structure analysis. Samples of silanes blended with photoiniator were further dissolved in $CDCl_3$ and then characterized using a INOVA 500 MHz nuclear magnetic resonance (NMR) spectrometer to obtain their ^{13}C- and ^1H-NMR spectra after they were cured by UV radiation.

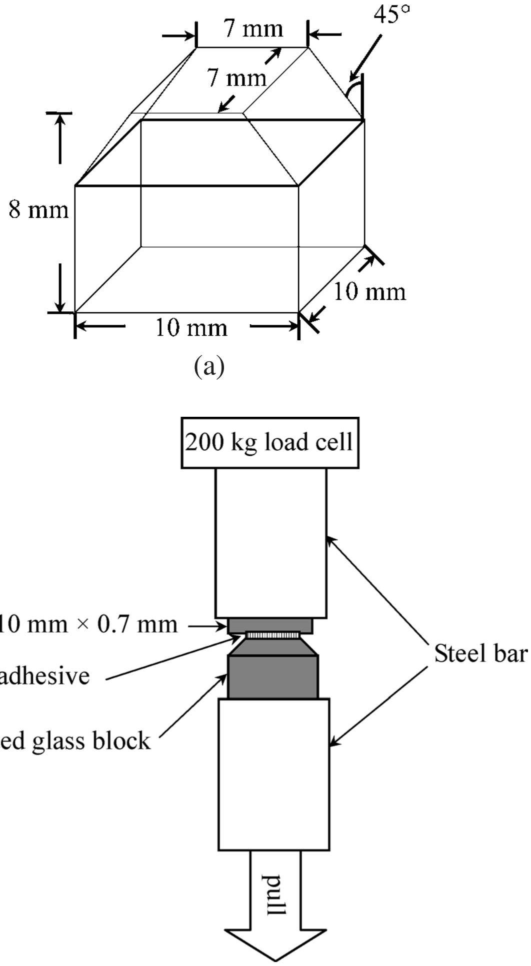


Figure 1. (a) The size of the truncated glass substrate and (b) the test scheme for adhesion strength measurement.

3. RESULTS AND DISCUSSION

3.1. Chemical reaction of silane containing alkoxy group

Most of the studies relative to the chemical reaction of silane containing alkoxy group ($-SiOCH_3$) have focused on the hydrolysis of alkoxy group to produce $-Si-OH$ [15, 16] in order to improve the adhesion of polymeric materials to inorganic surfaces. In this work, silanol was formed as alkoxy group of silane underwent UV irradiation. FT-IR and NMR analyses were utilized to explain the chemical reactions of silanes blended with photoinitiator. Figure 2 shows the FT-IR spectra of vinyl group silane and silane blended with photoinitiator after UV curing for 60 min. Prior to UV curing, the alkoxy group of silane presents a

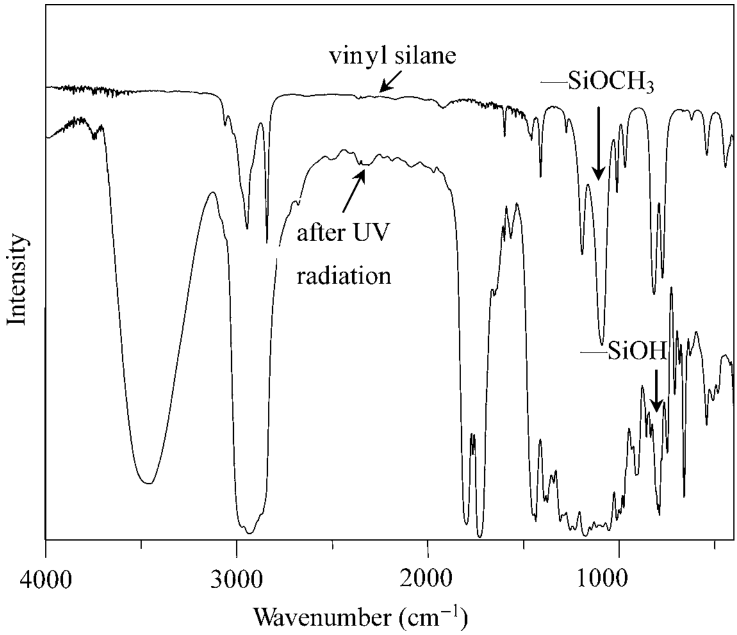


Figure 2. FT-IR spectra of vinyl group silane and silane blended with photoinitiator after UV curing for 60 min.

peak at 1088–1076 cm^{-1}. During UV curing, the alkoxy group of silane reacts with $H^+SbF_6^-$ to generate $-Si-OH$, as evidenced by the peak at 953 cm^{-1}. The other four silanes studied in this work exhibit the same result. On the other hand, a comparison of ^{13}C-NMR spectra for the specimens of vinyl group silane and the silane blended with photoinitiator subjected to UV curing shown in Fig. 3a and 3b reveals that the peak at $\delta = 50.437$ ppm, corresponding to alkoxy group of silane vanished after UV curing. Decomposition of alkoxy group of silane by cationic photoinitiator to form $-Si-OH$ has been reported elsewhere [17]. Also, the peak at $\delta = 3.569$ ppm in ^1H-NMR spectrum shown in Fig. 3c corresponds to H atoms on alkoxy groups of silane which disappeared when silane was blended and subsequently underwent UV curing, as shown in Fig. 3d.

Apparently, the Brønsted acid generated during UV curing attacks the alkoxy group of the silane and the acid converts the alkoxy groups into silanols, as depicted in equation (7). The ITO glass surface was treated electrochemically to generate hydroxyl groups [18–20] that react with silanol to produce hydrogen bonding, as shown in equation (8). This is similar to the hydrolysis of silane in order to enhance the adhesion to metals such as steel [21], copper [8] and aluminium [22].

$$\equiv Si-OR' \xrightarrow{H^+SbF_6^-} \equiv Si-OH \qquad (7)$$

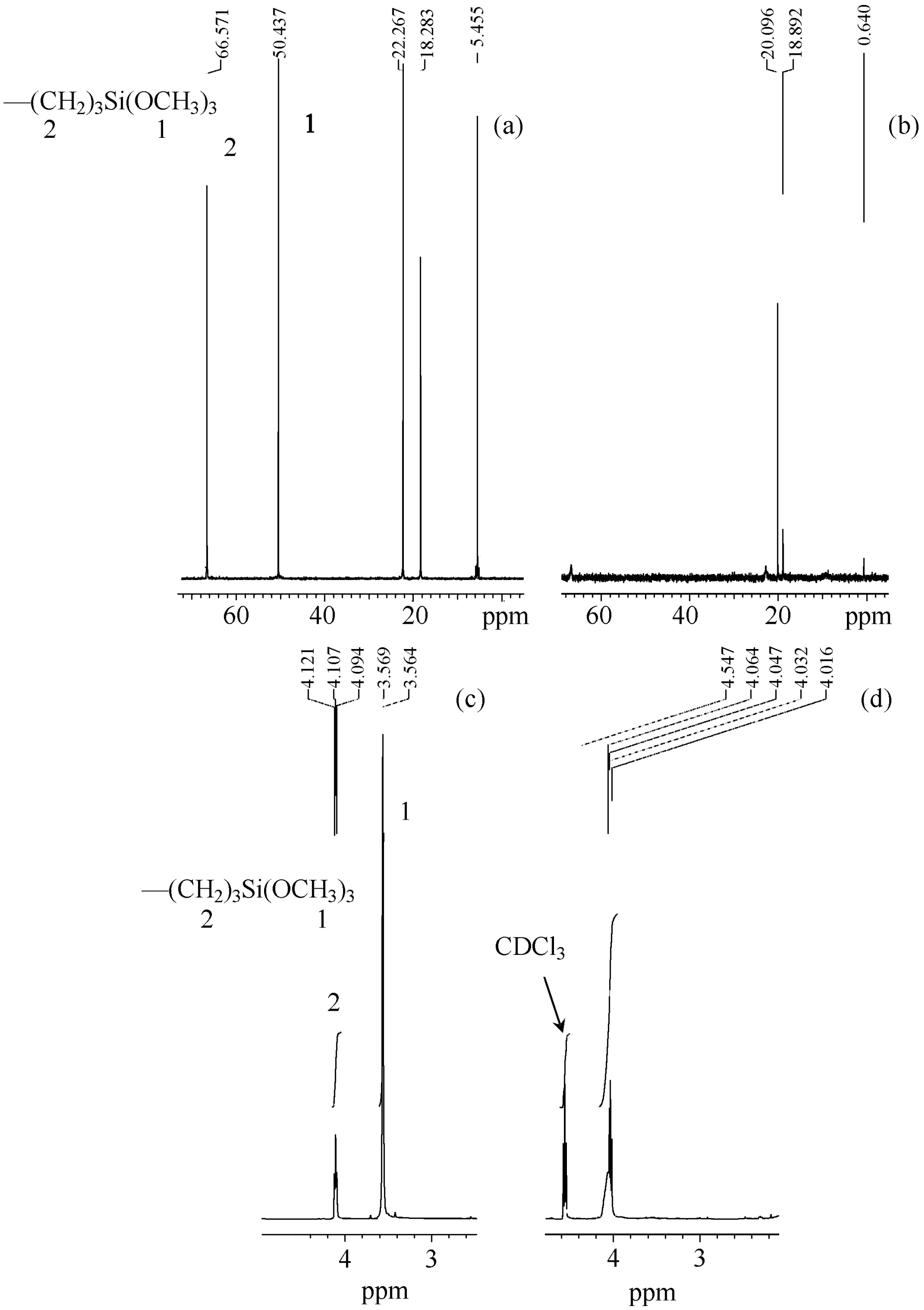
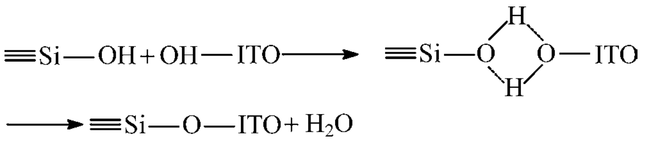


Figure 3. ^{13}C-NMR spectra of (a) the alkoxy group of vinyl silane and (b) vinyl silane blended with photoinitiator after UV curing. ^{1}H-NMR spectra of (c) the alkoxy group of vinyl silane and (d) vinyl silane blended with photoinitiator after UV curing.



$$\equiv\mathrm{Si}{-}\mathrm{OH}+\mathrm{OH}{-}\mathrm{ITO}\longrightarrow\ \equiv\mathrm{Si}{-}\mathrm{O}\overset{\mathrm{H}}{\underset{\mathrm{H}}{\diamond}}\mathrm{O}{-}\mathrm{ITO}\longrightarrow\ \equiv\mathrm{Si}{-}\mathrm{O}{-}\mathrm{ITO}+\mathrm{H_2O} \tag{8}$$

3.2. The reaction between organo-functional group of silane coupling agent and cationic photoinitiator

The reactions between organo-functional group of silane coupling agent and cationic photoinitiator affect the adhesion strength of adhesive resin to ITO glass. This work focused on the effect of UV curing time of the samples prepared by mixing 1 g of cationic photoinitiator and 0.5 g of selected silane coupling agent. Table 2 shows the UV curing times of different silanes blended with photoinitiator. The lengths of UV curing time were in the order: amino $>$ vinyl $>$ methacrylic $>$ acrylic $>$ epoxy. The UV curing time of epoxy silane is the shortest because a cationic photoinitiatior was used in this work. The silanes containing acrylic, methacrylic and vinyl groups can undergo free-radical polymerization. Different C=C double bond structures in these organo-functional groups imply different UV reactivities. The free-radical polymerization reaction was relatively slow in comparison to the cationic polymerization in the system due to the low production rate of initiating radicals during the photolysis of the onium salt [23]. Meanwhile, the UV reactivity was in the order: acrylic $>$ methacrylic $>$ vinyl $\approx$ allyl $>$ double bond in the main chain. The acrylic group silane exhibits the highest UV reactivity because its $-$COOR group is electrophobic [24]. It can decrease the density of electron cloud on the C=C bond and makes it easier to bond with the free radicals. The UV reactivity of methacrylic group silane is lower than that of acrylic group since it possesses electrophilic $-CH_3$ group [24] which increases the density of electron cloud. The vinyl group silane has the lowest UV reactivity because of the high density of electron cloud on the C=C bond and lack of substituent group. Different functional groups with the C=C double bond affect differently the cross-link density of the network and the length of curing time of resin [25–27]. As to the amino-functional

Table 2.
Curing times of samples containing various silanes

Silane type	Curing time (min)
Vinyl	60
Epoxy	5
Amino	(Uncurable)
Acrylic	8
Methacrylic	45

silane, it inhibits the UV curing reaction and thus resin sample could not be cured, as described by Plueddemann [28].

From the ^{13}C-NMR spectra shown in Fig. 4a and 4b, the peaks of epoxy group silane at $\delta = 50.269$ and 43.885 ppm disappeared after UV curing. There are two signals appearing in peak 2 of Fig. 4a and their locations are too close to be differentiated as to which one is methoxy carbon and which one is epoxy carbon. However, the peak at 43.885 ppm corresponds to epoxy carbon and it disappeared after UV irradiation, which means epoxy silane had reacted with $H^+SbF_6^-$. For the ^{1}H-NMR spectra shown in Fig. 4c and 4d, the peaks of epoxy group silane at $\delta = 2.6$–3 and 3.2–3.5 ppm disappeared after UV curing, and the peaks at $\delta = 4.8868$ and 4.046 ppm are generated by the photoinitiator. The reaction of epoxy silane with $H^+SbF_6^-$ produces oxiranium ion as depicted in equation (9) [29].

According to equation (1), the phenyl radical was produced by the photoinitiator during UV irradiation and the radical would bond to the double bonds present in the monomer [30]. Organfunctical groups of vinyl, acrylic and methacrylic silanes all possess C=C double bonds which can undergo free-radical polymerization as described by equations (10) [31], (11) [32] and (12) [33]. The chemical structures for the three types of silanes, vinyl, acrylic and methacrylic, were evidenced by the ^{1}H-NMR spectra shown in Figs 5a, 6a and 7a, respectively. The peaks of double bonds of vinyl, acrylic and methacrylic groups are at $\delta = 6.125$–6.022 and 5.944–5.873 ppm, $\delta = 6.4$–6.1 and 5.827–5.928 ppm and $\delta = 6.091$ and 5.941 ppm, respectively. After UV curing, the C=C double bonds of these three types of silanes reacted with phenyl radical and hence the corresponding peaks disappeared as presented in Figs 5b, 6b and 7b, respectively. Furthermore, the H atoms on the phenyl group of the photoinitiator result in the peak at $\delta = 7$–8 ppm.

Figure 8 shows ^{1}H-NMR spectra of the amine group of silane and the silane blended with photoinitiator after UV curing. A comparison of Fig. 8a and 8b reveals the disappearance of the peak at $\delta = 2.696$ ppm. The amine group of silane is a Lewis base and the nitrogen atom of the amine with a lone pair of electrons might react with Brønsted acid photoinitiator to produce ammonium salts, as depicted in equation (13) [34]. The production of ammonium salt would inhibit the photocuring process of adhesive resin [28]. On the other hand, the peaks at $\delta = 4.8868$ and 4.046 ppm are generated from the photoinitiator as, shown in Fig. 8b.

Figure 9a–f shows morphology of epoxide resin containing various types of silanes after UV curing and the degree of shrinkage can be readily seen from the morphology change of specimen surfaces. Referring to Fig. 9 and Table 2, it is found that a shorter curing time implies a larger shrinkage and *vice versa*. As shown in Fig. 9a, the resin containing vinyl group exhibits the smoothest surface with the smallest degree of shrinkage and the longest curing time. The degree of shrinkage was found to be in the order: vinyl $<$ methacrylic $<$ acrylic $<$ epoxy $<$ sample free of silane additive $<$ amino.

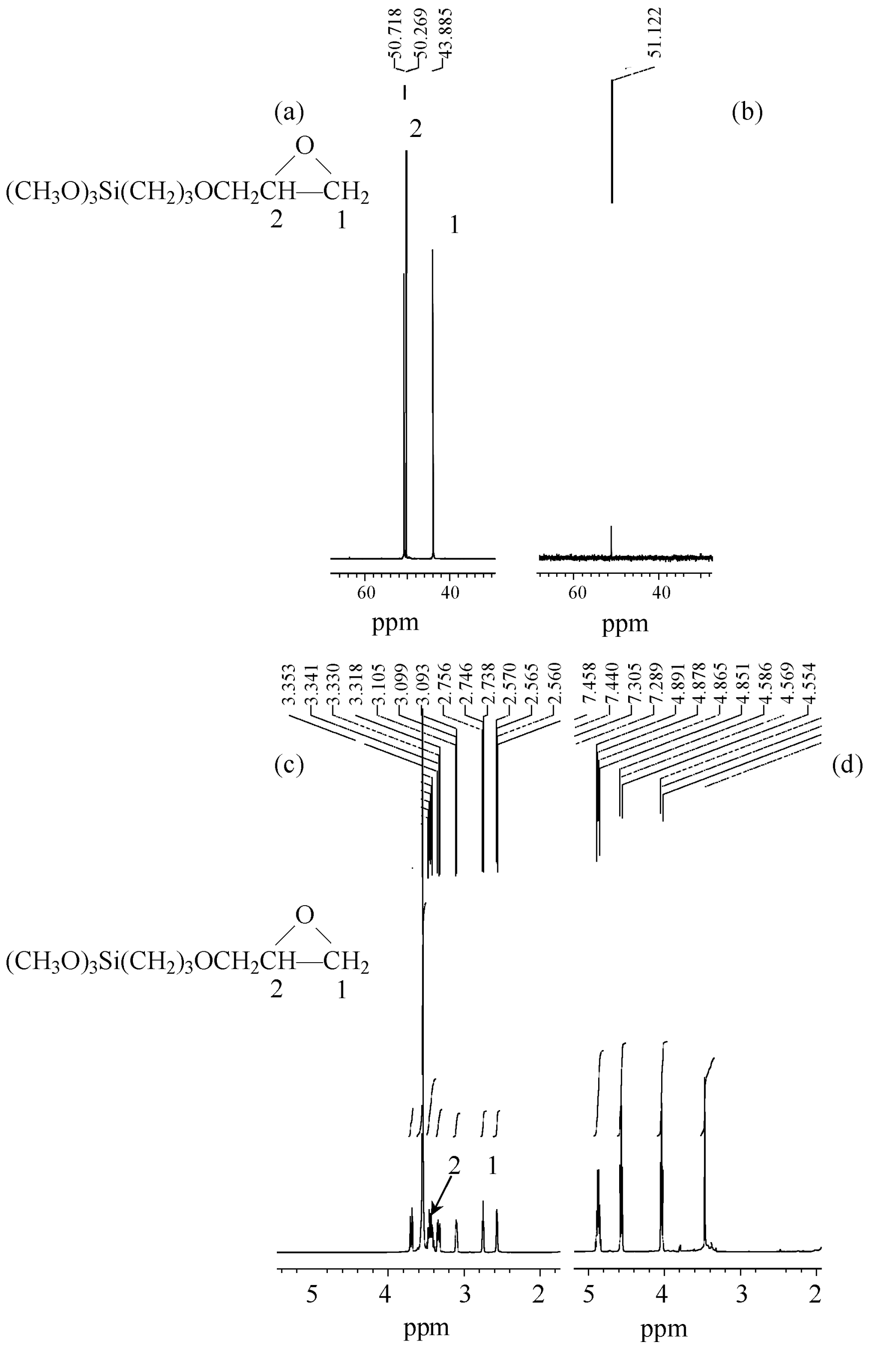


Figure 4. ^{13}C-NMR spectra of (a) the epoxy silane and (b) epoxy silane blended with photoinitiator after UV curing. ^{1}H-NMR spectra of (c) the epoxy silane and (d) epoxy silane blended with photoinitiator after UV curing.

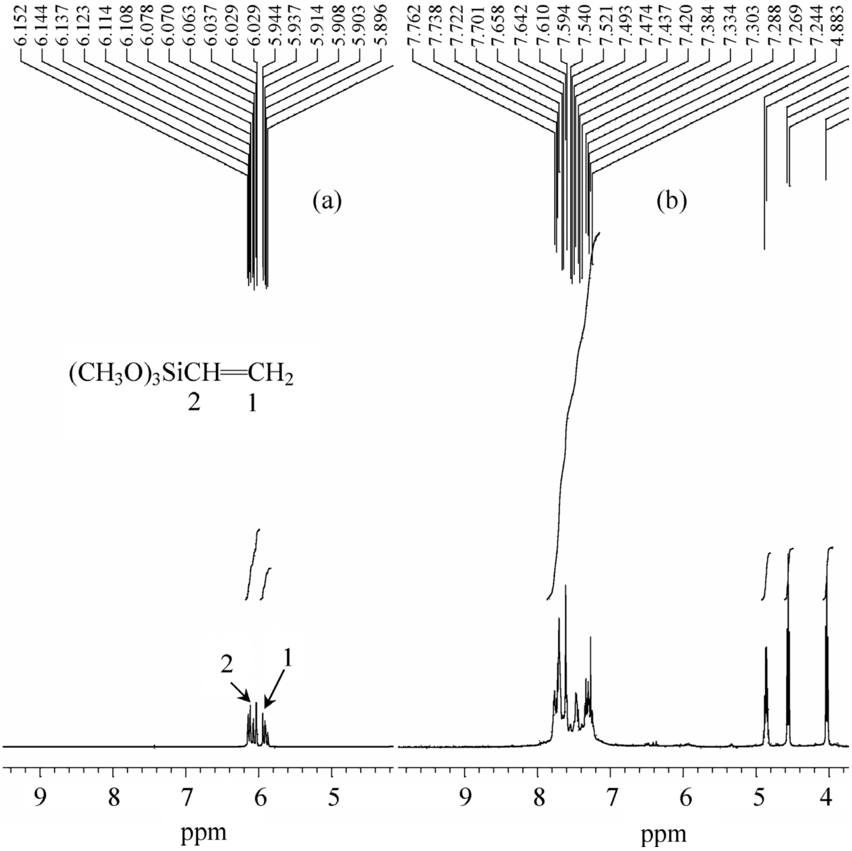
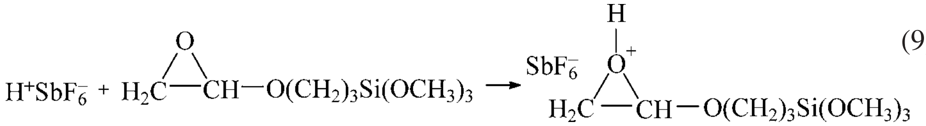


Figure 5. ^{1}H-NMR spectra of (a) vinyl silane and (b) vinyl silane blended with photoinitiator after UV curing.



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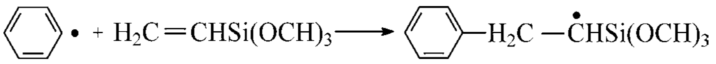

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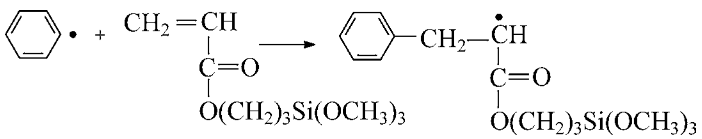

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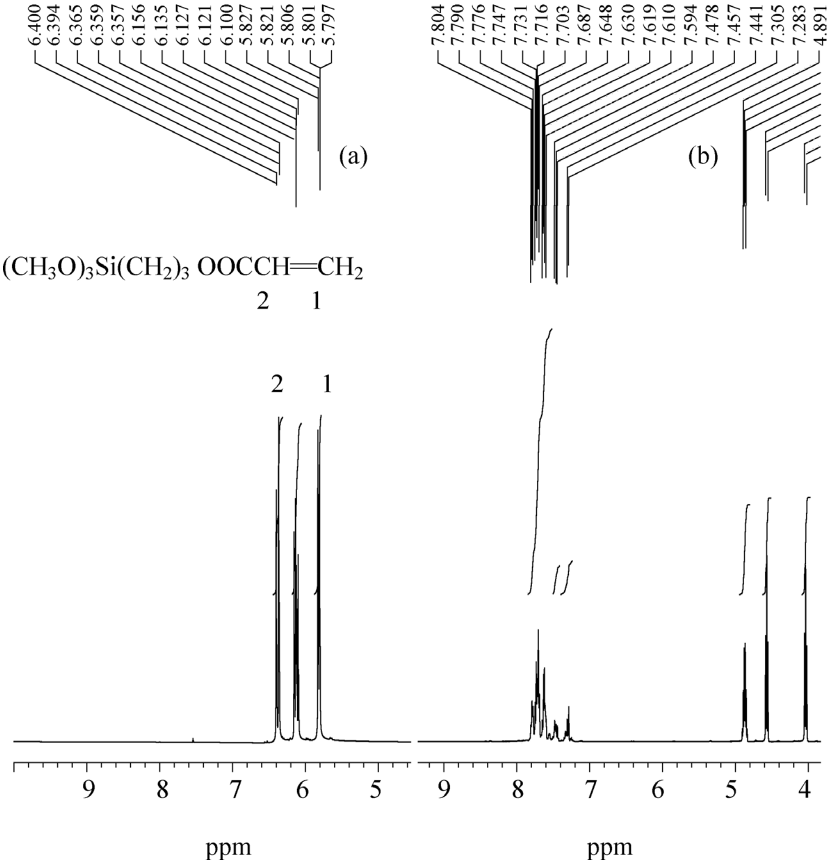
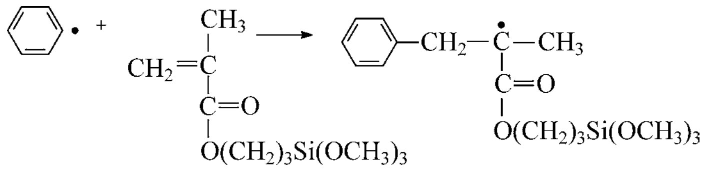


Figure 6. ^{1}H-NMR spectra of (a) acrylic silane and (b) acrylic silane blended with photoinitiator after UV curing.



(12)

$$H^+SbF_6^- + H_2\ddot{N}(CH_2)_3Si(OCH_3)_3 \longrightarrow SbF_6^-\ H_3\overset{+}{N}(CH_2)_3Si(OCH_3)_3 \tag{13}$$

3.3. Effect of functional group type of silane on adhesion strength

Shrinkage of the resin occurring during the curing process may affect its adhesion strength because it may induce internal stress that pulls the adhesive resin away from substrate [35, 36]. Figure 10 shows the adhesion strengths of five adhesive resins blended with different type of silanes and resin without silane evaluated by a

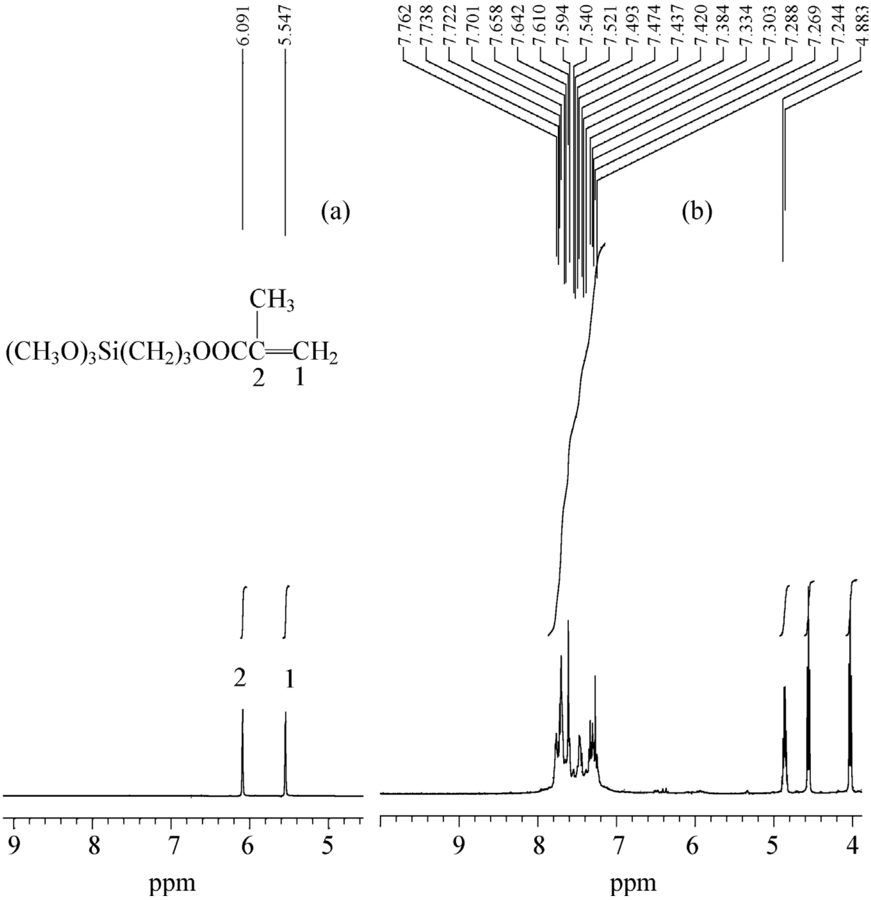


Figure 7. ^{1}H-NMR spectra of (a) methacrylic silane and (b) methacrylic silane blended with photoinitiator after UV curing.

pull test. The adhesion strength to ITO glass was found to be in the order: vinyl > methacrylic > acrylic > epoxy ≈ sample free of silane additive > amino and the values are 79.77, 78.18, 59.39, 36.78, 35.57 and 22.77 kg/cm^2, respectively. The trend of adhesion strengths is similar to that reported by Priola *et al.*, who studied the adhesion of BGEDA resins containing vinyl, methacrylic and acrylic silanes to steel sheet [21]. The results of adhesion study also evidenced that a reduction in resin shrinkage implied a decrease of internal stress that would benefit the adhesion strength of resins.

3.4. Effect of weight percentage of vinyl group silane on adhesion strength

According to the results of pull test, the adhesive resin containing vinyl group silane exhibited the highest adhesion strength. Subsequent study was hence focused on the adhesion property to ITO glass for such resin containing different weight percentages of vinyl group silane. From the results of pull test shown in Fig. 11,

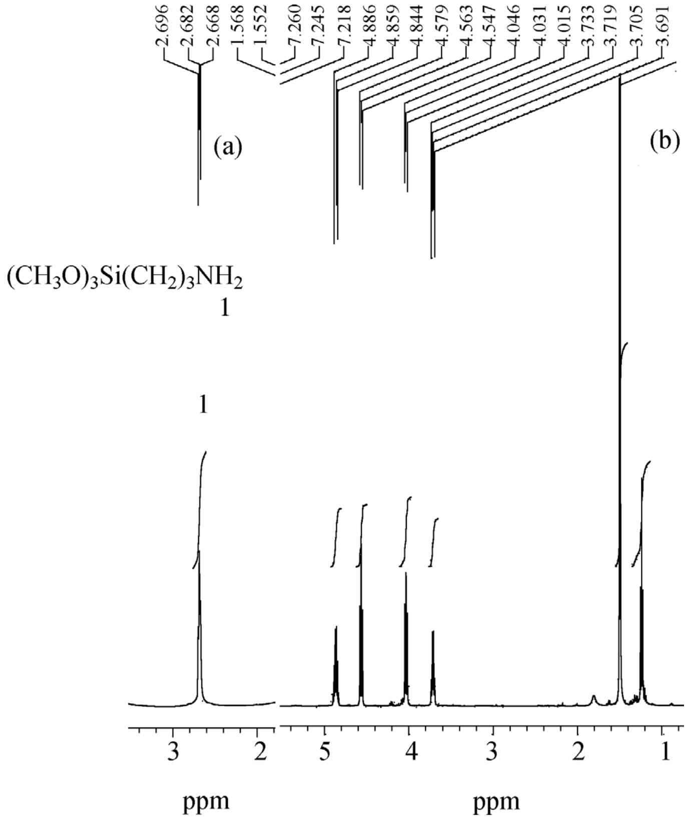


Figure 8. ^{1}H-NMR spectra for (a) amino silane and (b) amino silane blended with photoinitiator after UV curing.

one can readily see that the resin sample containing 1 wt% of vinyl group silane possesses the highest adhesion strength, 91.42 kg/cm^2. When the weight percentage of silane was further increased, a decreasing trend in adhesion strength of the resin was observed. Silanol, produced from the UV-irradiated alkoxy, reacts with the hydroxyl groups of ITO glass to generate Si—O—ITO bonds and some H_2O as shown in equation (8). Meanwhile, Si—O—Si bonds and H_2O are generated from the reaction between silanol groups [37, 38]. As a result, there was more H_2O generated *via* these reactions when more silane was added in the resin. As a matter of fact, the addition of 1 wt% of silane giving the highest adhesion strength among all of the samples is in good agreement with the report of Gupta and Dhuldhoya [39]. Furthermore, an excessive amount of silane might dilute the concentration of resin and decrease the adhesion strength, which was reported by Gu *et al.* who studied the adhesion of epoxide resin containing γ-aminopropyltrimethoxysilane [8].

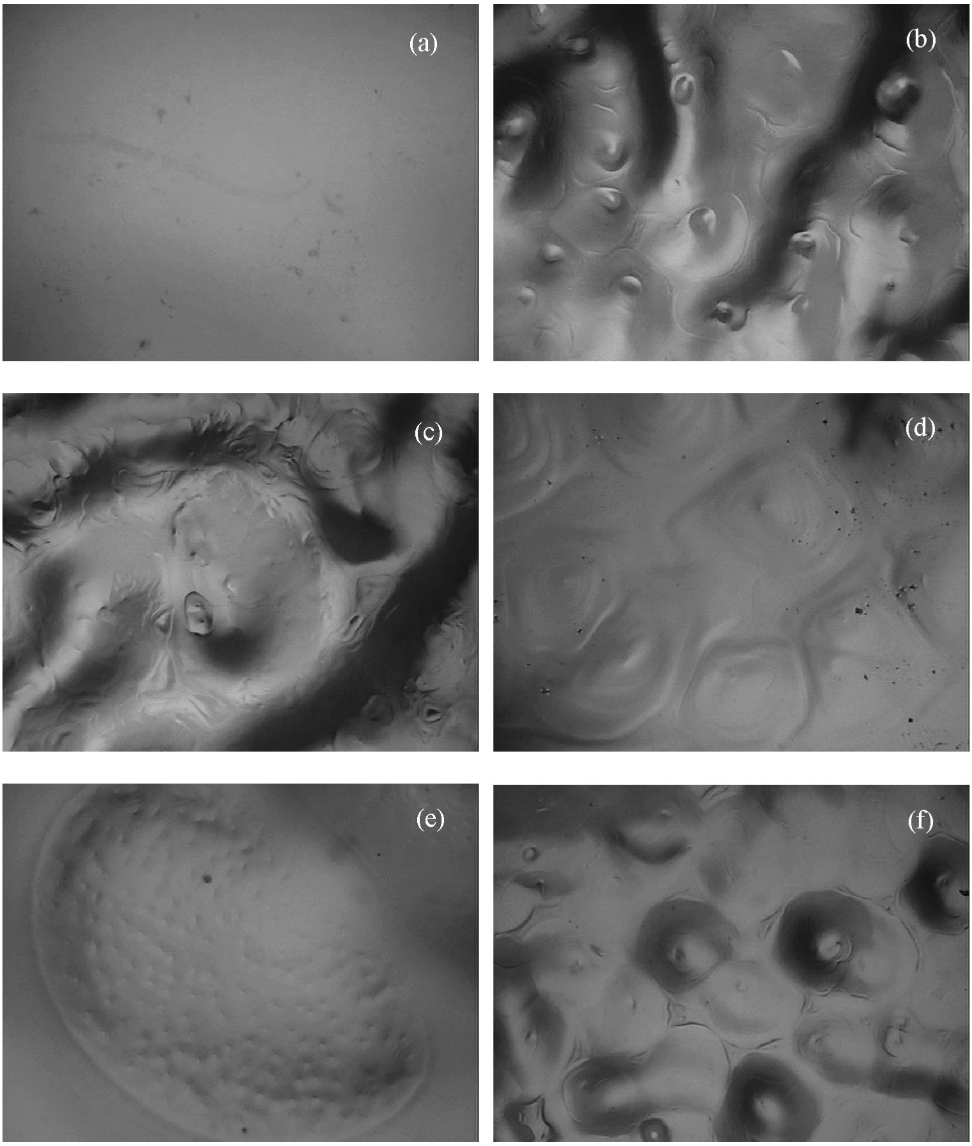


Figure 9. Optical microscope images (20× magnification) for epoxide resin containing (a) vinyl, (b) epoxy, (c) amino, (d) acrylic, (e) methacrylic slianes and (f) free of silane additive after UV curing.

4. CONCLUSIONS

In this study we investigated the reactivity and adhesion property to ITO glass of epoxide resins containing different types of organo-functional silanes. The epoxide resins containing different organo-functional silanes reacted with photoinitiator *via* different reaction mechanisms and hence resulted in different UV curing times. It was found that a longer curing time resulted in less shrinkage of the resin after

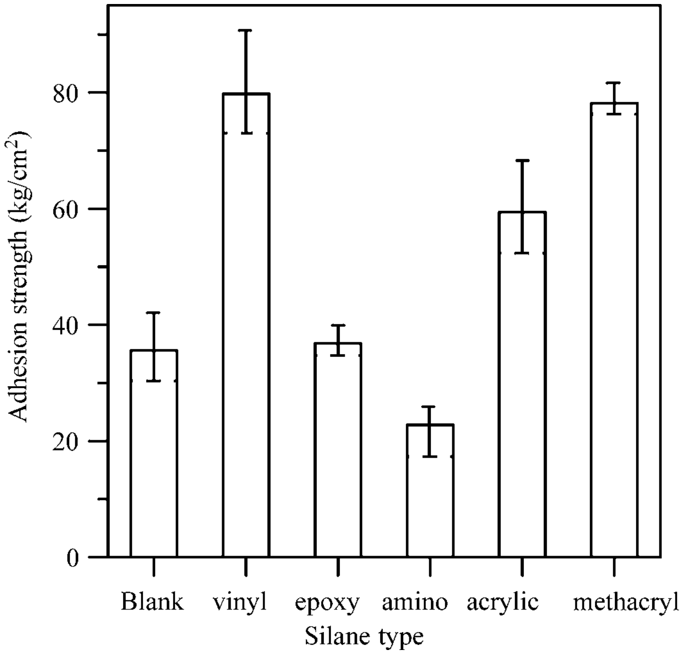


Figure 10. Adhesion strength of epoxide resins containing various types of silanes to ITO glass.

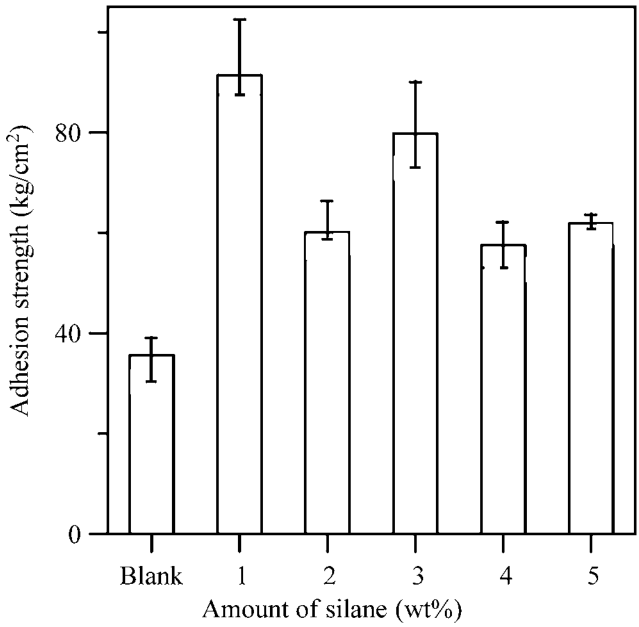


Figure 11. Adhesion strength of epoxide resins containing different weight percentages of vinyl silane to ITO glass.

UV curing. A low degree of shrinkage induced low internal stress in the cured resin and hence resulted in high adhesion strength. The pull test results showed that the adhesion strength of resins containing silanes to ITO glass substrate was in the order: vinyl $>$ methacrylic $>$ acrylic $>$ epoxy $\approx$ sample free of silane additive $>$ amino. The enhancement of adhesion property resulting from the addition of vinyl group silane in epoxide resin was attributed to the promotion of free-radical polymerization with photoinitator. The resin sample containing 1 wt% of vinyl group silane exhibited the highest adhesion strength of 91.42 kg/cm^2 to ITO glass. Further increase of silane content in the resin would generate too much H_2O that would dilute the resin concentration and thus deteriorate its adhesion strength.

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