Synthesis and Characterization of Alkali-Soluble Photosensitive Polysiloxane Urethane Acrylate

Fang Sun,[1,2] Bo Liao,[3] Lu Zhang,[1,2] Hong-Guang Du,[1,2] Yue-Dong Huang[4]

[1]State Key Laboratory of Chemical Resource Engineering, Beijing University of Chemical Technology, Beijing 100029, People's Republic of China
[2]College of Science, Beijing University of Chemical Technology, Beijing 100029, People's Republic of China
[3]Second Institute, Equipment Academy of Second Artillery, Beijing 100085, PR China
[4]Department of Chemical Engineering, Jilin Vocational College of Industry and Technology, Jilin 132013, PR China

ABSTRACT: A novel alkali-soluble photosensitive polysiloxane urethane acrylate (APSUA) was synthesized via reacting bifunctional isocyanate with three kinds of hydroxyl group respectively, from hydroxyl silicone oil, dimethylol propionic acid (DMPA), and a compound which also has acrylate groups. The compound was prepared via reacting acrylic acid with N,N,N',N'-tetraglycidyl -4,4'-diamino diphenyl methane (AG-80). The optimum temperature for the synthesis reaction of APSUA was found to be 50°C. The structure of the oligomer was characterized by IR, ^{1}HNMR, and GPC. The number-average molecular weight of the APSUA was 7795. The APSUA possesses excellent compatibility with most of acrylate monomers. The effect of concentration of photoinitiator and monomers on the photopolymerization kinetics of the oligomer APSUA was investigated by real-time infrared spectroscopy (RT-IR). The results show that APSUA can well photopolymerize under UV-irradiation. The optimal concentration of photoinitiator (Darocur 1173) was determined as 0.1 wt %. The system of APSUA with DPGDA monomer has higher polymerization rate (0.0521 s^{-1}) and higher double bond conversion (93.2%), respectively. The APSUA system with acrylate monomers can form regular image under UV-irradiation through a patterned mask. The cured coating film of the APSUA system possesses excellent flexibility, toughness and heat resistance. The APSUA system can dissolve in 1% sodium carbonate solution easily and completely. © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 120: 3604–3612, 2011

Key words: alkali-soluble; polysiloxane; photosensitive; solder resist ink

INTRODUCTION

Photosensitive polysiloxane plays a very important role in the field of photopolymerization materials for good resistance to high temperature, good flexibility,[1–5] environmental friendly, energy saving,[6–11] improved workability, etc. In recent years, as a higher packaging density has been required, photosensitive polysiloxane has been gradually used for the solder resist ink in the field of manufacture of flexible printed circuit boards. The outstanding advantages of photosensitive polysiloxane compared with conventional solder resist ink are as follows: sufficient flexible, folding resistance, and excellent heat resistance. However, to form accurate pattern, a large amount of organic solvent as developer is used during developing process, which leads to severe environmental pollution. Therefore, in the past 2 decades much research work has been done to replace organic solvent. Many investigators have studied and developed alkali-soluble or water-soluble photosensitive polymer to diminish pollution,[12–15] such as base-dissolved acrylated polyester and water-soluble photosensitive epoxy phenolic resin, but there is no report on alkali-soluble photosensitive polysiloxane used for the solder resist ink at present. Poor compatibility of photosensitive polysiloxane with other resins and acrylic monomers limits its application in UV-curable materials.[16–18]

The purpose of this article is to design and synthesize an environmental friendly dilute alkali-soluble photosensitive polysiloxane for the solder resist ink, which is also compatible with acrylic monomers well. The alkali-soluble photosensitive polysiloxane urethane acrylate (APSUA) was synthesized from hydroxyl-terminated polysiloxane, isophorone diisocyanate (IPDI), dimethylol propionic acid (DMPA), acrylic acid (AA), and N',N'-tetraglycidyl -4,4'-diamino diphenyl methane (AG-80) as raw materials. The structure of alkali-soluble photosensitive polysiloxane urethane acrylate (APSUA) was characterized by ^{1}HNMR, IR and GPC. The
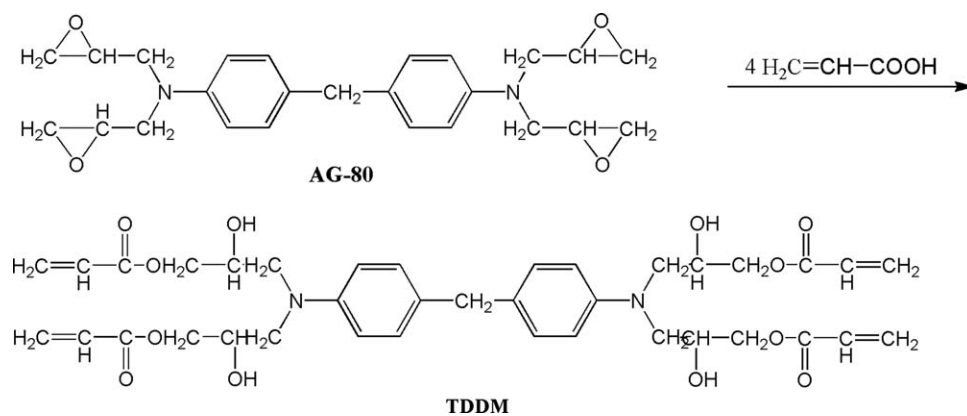
Scheme 1 Synthesis reaction equation of N,N,N',N'-tetra(2-hydroxypropyl acrylate)-4,4'-diamino diphenyl methane (TDDM).

photo-polymerization properties were researched by real time infrared spectroscopy (RT-IR). The compatibility of the APSUA system, adhesion, hardness and heat resistance of the APSUA cured film were also investigated.

EXPERIMENTAL

Materials

Hydroxyl-terminated polysiloxane (Q4-3667) was obtained from Dow Corning Corp. Dimethylol propionic acid (DMPA, AR), acrylic acid (AA, AR) and epoxy resin N,N,N',N'-tetraglycidyl-4,4'-diamino diphenyl methane (AG-80) were provided by Beijing Yili Fine Chem Co. Isophorone diisocyanate (IPDI) was obtained from Qingdao Xinyutian Chemical Co. Dipropylene glycol diacrylate (DPGDA), 2-hydroxyethyl acrylate (HEA), isobornyl acrylate (IBOA) and trimethylol propane triacrylate (TMPTA) were purchased from Beijing Dongfang Chemical Co. Photoinitiators 2-hydroxyl-2-methyl-1-phenyl propane-1-one (Darocur 1173) and polyurethane acrylate OAK-27 was obtained from Ciba Geigy Co. Dibutyltin dilaurate (DBTDL) was supplied by Shanghai Chemical Reagents Co.

Synthesis

Synthesis of N,N,N',N'-tetra(2-hydroxypropyl acrylate)-4,4'-diamino diphenyl methane (TDDM)

TDDM was synthesized in terms of Scheme 1. N,N,N',N'-tetraglycidyl-4,4'-diamino diphenyl methane (0.56 g, 0.005 mol) and acrylic acid (1.44 g, 0.02 mol) were dissolved in N, N-dimethyl formamide (DMF) and reacted at 90°C under stirring for 5 h until the characteristic peak of epoxy group in the infrared spectra disappeared. The solvent was removed by reduced pressure distillation, and the product was purified by column chromatography (hexane/ethyl acetate = 4 : 1). The remainder was a

brown viscous liquid and the yield was approximately 82%.

IR (KBr, cm^{-1}): 3325–3576 cm^{-1} (—OH), 2857–2957 cm^{-1} (—CH$_3$, —CH$_2$—), 1739 cm^{-1} (>C=O), 1415–1615 cm^{-1} (>C=C<), 1514–1615 cm^{-1}(⟨◯⟩).

^{1}H-NMR (CDCl$_3$, ppm): 7.04 (⟨◯⟩—H), 6.52 (—C(=O)—C=C<), 5.95 (—C(=O)—C=C<), 4.43 (—◯—CH—◯—), 3.67 (—C(=O)—O—C<), 2.38 (>C—C(=O)—C<), 1.95 (>C—CH—N<).

Synthesis of alkali-soluble photosensitive polysiloxane urethane acrylate (APSUA)

The APSUA was synthesized according to the following procedure. First, Q4-3667 (40 g, 0.02 mol) was added in a four-necked flask equipped with a mechanical stirrer, a thermometer and a cooler, then IPDI (4.44 g, 0.02 mol) was added dropwise over 30 min into the flask. The reaction mixture was stirred at 50°C in the presence of DBTDL as a catalyst until the value of isocyanate reached to the theoretical one of mono-isocyanate by dibutylamine titration of NCO groups[19] namely, value of isocyanate reached to half of initial value. Subsequently, dimethylol propionic acid (DMPA, 2.42 g, 0.02 mol) was added into the flask and the reaction mixture was stirred until the absorption peak of —NCO group in the infrared spectra disappeared. Then, IPDI (4.44 g, 0.02 mol) was added into the mixture. When the value of isocyanate reached to half of initial value (based on second added IPDI), TDDM was added in. The reaction was stopped until the absorption peak of —NCO group in the infrared spectra completely disappeared. The product was purified by column chromatography (hexane/ethyl acetate = 3 : 1). The yield of APSUA was approximately 73%. The synthesis process of APSUA is shown in Scheme 2.

IR (KBr, cm^{-1}): 3324 cm^{-1}, 1534 cm^{-1} cm^{-1} (N—H), 2904 cm^{-1}–2965 cm^{-1} cm^{-1} (C—H), 1717 cm^{-1} (C=O),
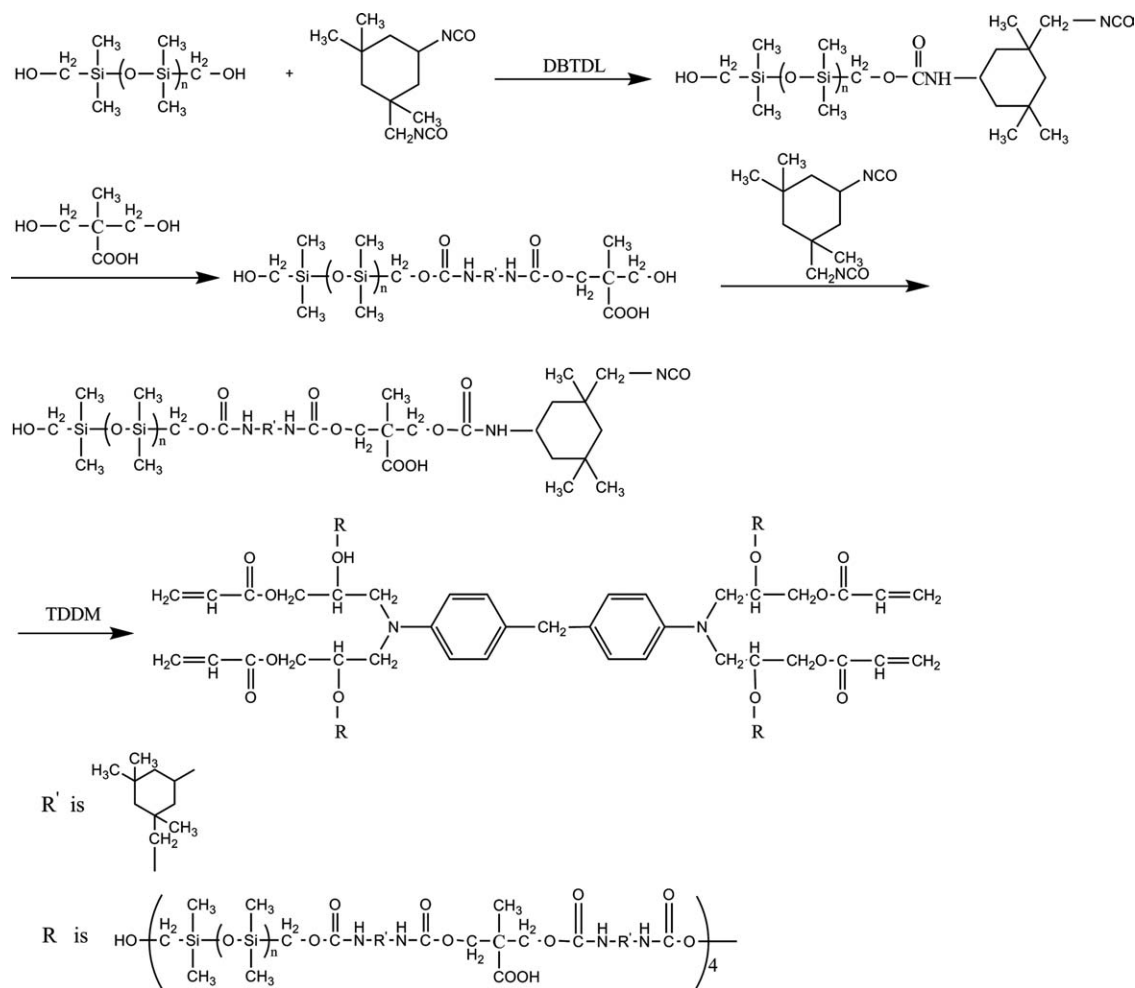
Scheme 2 Synthesis reaction equation of alkali-soluble photosensitive polysiloxane urethane acrylate (APSUA).

1102 cm^{-1} (Si—O—Si), 1266 cm^{-1} and 804 cm^{-1} (Si—CH$_3$), 14,535–1534 cm^{-1} (>C=C<), 1534–1640 cm^{-1}(◯).

^{1}HNMR (CDCl$_3$, ppm): 0.062 (—Si—C—H), 2.42 (◯—H), 4.20 (◯—C—◯—), 6.42 (—C—C=C), 6.52 (—C—C=C), 1.98 (—C—C—C—), 1.03 (—C—C—N), 7.27 (◯—H), 3.65 (—C—N—C—, —C—O—C—), 2.63 (—C—N—C—), 1.67 (◯—C—).

Characterization of APSUA

The FTIR spectra were obtained on a Nicolet 50XC spectrometer (Nicolet, USA) and scanned between 400 and 4000 cm^{-1}.

^{1}HNMR spectra were recorded on an AV600MHz (Bruker, USA) spectrometer, using CDCl$_3$ as the solvent and tetramethylsilane as an internal standard.

The molecular weight of oligomer was determined by Water 515–2410 gel permeation chromatography (GPC, Water, USA). Tetrahydrofuran, 1.0 mL min^{-1}, was used as the mobile phase. Calibration was carried out by linear polystyrene of known molecular weight and dispersity. Thermal stability was determined on STA-449C simultaneous thermal analyzer (Netzsch, Germany). Samples were run from 30 to 500°C with a heating rate of 10°C/min. Dynamical thermal mechanical analyses (DMTA) were performed on DMTA-IV (Rheometric Scientific Co., USA). The tensile properties of the tensile properties of APSUA film were measured using a material testing instrument (Instron-1211, USA) at 25°C. The rate of extension was 10 mm/min. Pencil hardness apparatus AR015 (Tianjing Instrument Co., China) was employed to measure the hardness of APSUA cured films.

Photopolymerization

The systems of APSUA with all kinds of monomers were employed as the photopolymerizable resins, varying concentration of monomers and the newly synthesized APSUA. All samples were investigated by real-time infrared spectroscopy (RTIR).
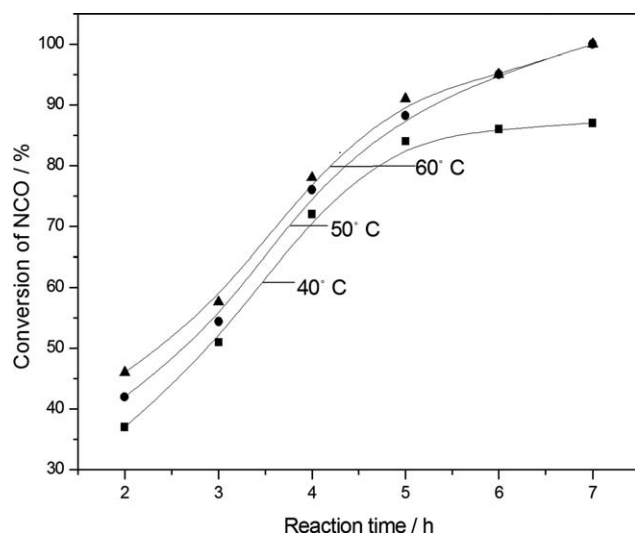
Figure 1 Effect of reaction temperature on the synthesis reaction of APSUA.

The basic principle of this new method of kinetic analysis consists in exposing the sample simultaneously to the UV laser beam, which induces the polymerization, and to the analyzing IR beam, which monitors the resulting drop in the absorbance of the reactive double bond. The IR spectrophotometer must be set in the absorbance mode and the detection wavelength fixed at a value where the monomer double bond exhibits a discrete and intense absorption, e.g., at 6100.70–6222.50 cm^{-1} for acrylic monomers. Upon laser irradiation, the fast decrease of the 6100.70–6222.50 cm^{-1} peak will accurately reflect the extent of the polymerization process since the absorbance increment. $(A_{6100-6222})_o$ - $(A_{6100-6222})_t$ is directly proportional to the number of acrylate functions which have polymerized.[20]

RTIR with a horizontal sample holder (Nicolet5700, Thermo Electron, USA, equipped with an extended range KBr beam-splitter and an MCT/A detector) was used to monitor the extent of polymerization. Photopolymerization was carried out at room temperature. The mixture of APSUA, monomers, and photoinitiators was placed in a mold made from glass slides and spacers with 15 ± 1mm in diameter and 1.2 ± 0.1 mm in thickness. A horizontal transmission accessory (HTA) was designed to enable mounting of samples in a horizontal orientation for FTIR measurements. A UV spot light source (EFOS Lite, Canada) was directed to the sample with light intensity of 10 mW/cm^2 (Honle UV meter, Germany). The decrease of the =C—H absorption peak area from 6100.70 to 6222.50 cm^{-1} in the near-IR range accurately reflects the extent of the polymerization since the change of the absorption peak area was directly proportional to the number of the (meth) acrylate

double bond that had polymerized. After baseline correction, conversion of the functional groups could be calculated by measuring the peak area at each time of the reaction and determined as the following:

$$DC\ (\%) = (A_0 - A_t) \times 100/A_0 \quad (1)$$

where DC is the degree of (meth) acrylate double bond conversion at t time, A_0 is the initial peak area before irradiation and A_t is the peak area of the double bonds at t time. The rate of photopolymerization is calculated by the differential of conversion of double bond versus irradiation time.[21]

Each sample was repeated three times and the error on the reported double bond conversion as a function of polymerization time was less than 2%. And in most case, it was less than 1%.

Property of photo-imaging

APSUA and DPGDA were mixed with a weight ratio of 60/40, subsequently, 1173 of 0.07% was added in the mixture to form a stock photosensitive liquid. The photosensitive liquid was coated onto a clear glass substrate and was covered with a patterned mask. Then, the photosensitive liquid layer was exposed for 90 s under a high-pressure mercury lamp (1000 W) through the patterned mask. Subsequently'the cured layer was developed in 1%Na$_2$CO$_3$ solution and a resist pattern was formed.

RESULTS AND DISCUSSION

Effect of reaction temperature on the synthesis of APSUA

Figure 1 shows the effect of reaction temperature on the reaction of isocyanate group with hydroxyl group. The conversion of NCO at 50°C increased about 15% over that at 40°C; however, it is very
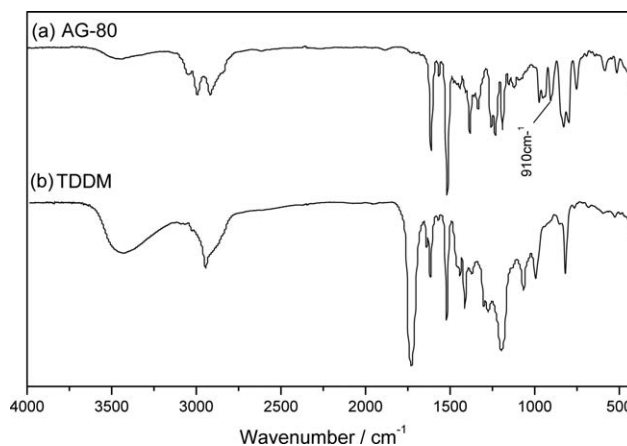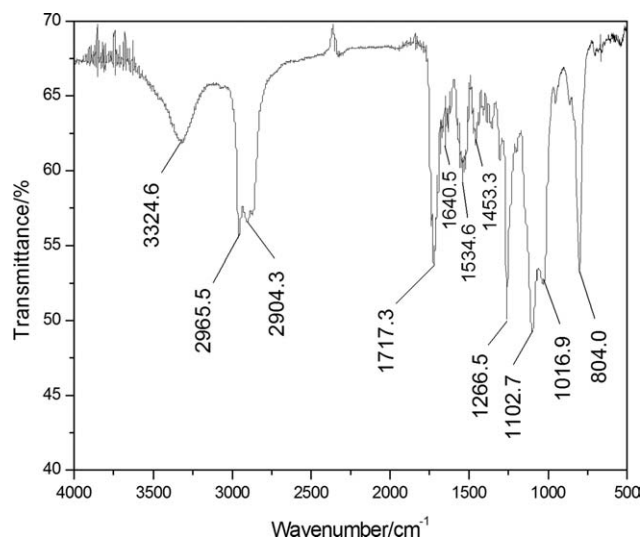

Figure 2 IR spectra of AG-80 and TDDM.

Figure 3 IR spectra of APSUA.

close to that at 60°C after reacting for 7 h. It thus proved that the rise of temperature can enhance the reaction rate. And higher temperature like 70°C or 80°C would easily result in gelation, mainly because it is hard to remove the reaction heat.

IR and ^{1}HNMR

IR of TDDM

The IR spectra of TDDM are shown in Figure 2. The absorption peak of epoxy group (910 cm^{-1}) group in the infrared spectra completely disappeared, indicating that the ring-opening reaction was accomplished.

IR of APSUA

The reaction of synthesized APSUA was also confirmed by IR spectra (Fig. 3). The characteristic absorption peaks of N—H, —O—C=O, Si—O—Si and Si—CH$_3$ appear at 3324 cm^{-1}, 1717 cm^{-1}, 1102 cm^{-1}, 1259 cm^{-1}, and 804 cm^{-1}, respectively, indicating that urethane, DMPA and polysiloxane were successfully linked with the N, N, N′, N′ -tetra(2-hydroxypropyl acrylate) -4, 4′- diamino diphenyl methane(TDDM).

^{1}HNMR of APSUA

The ^{1}HNMR spectra of the APSUA are shown in Figure 4. The different hydrogen atoms in APSUA molecule are shown in Scheme 3. From Figure 4 and Scheme 3, it is observed that the chemical shift of H3, H10, and H13 moved to 3.6 ∼ 3.8 ppm after the reaction, resulting from change in their chemical environment. However, the chemical shift of H7 in methylene connected to two benzene rings hardly changed before and after the reaction. Therefore, the structure of synthesized APSUA was determined by calculating the peak area ratio of H3, H10, and H13 to the H7 in ^{1}HNMR spectra of APSUA. The calculated ratio was 15.56 : 1, which is close to theoretical value (17 : 1), indicating the structure of synthesized APSUA nearly corresponds with the idea structure shown in Scheme 2.

GPC of TDDM and APSUA

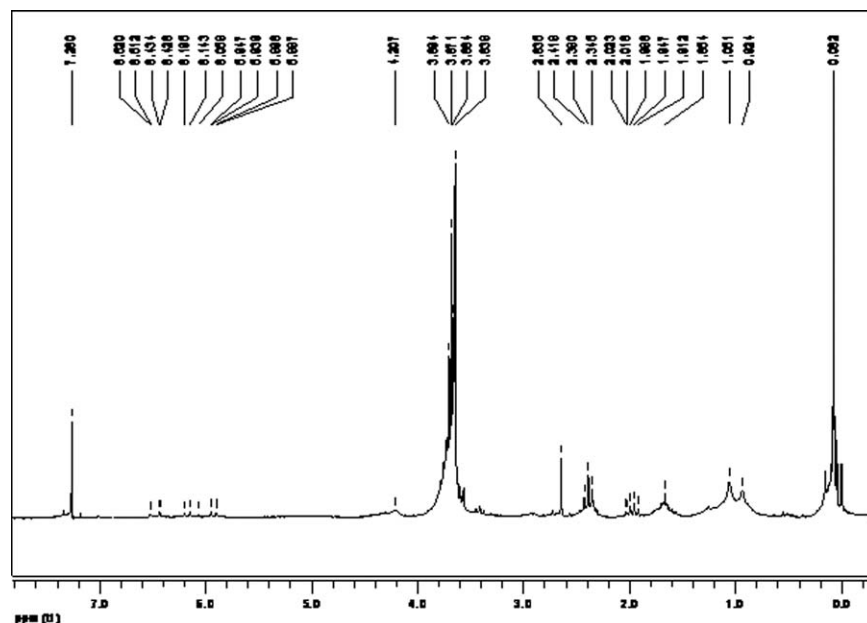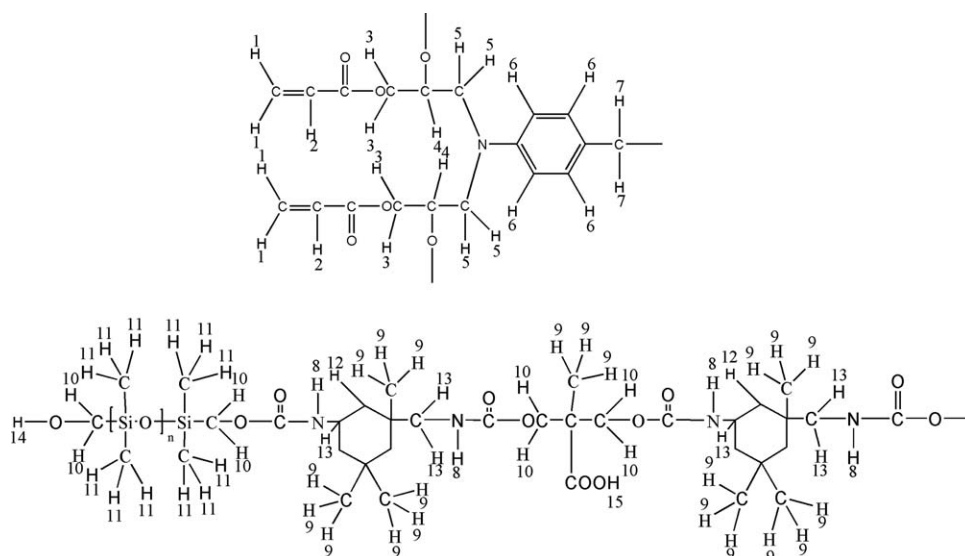The molar masses and molar mass distributions of the synthesized TDDM and APSUA were analyzed

Figure 4 ^{1}HNMR spectra of APSUA.

Scheme 3 Different H in APSUA molecule.

by GPC and the results were shown in Table I. The GPC instrument was calibrated using linear polystyrene standard. The values of molar masses of TDDM, which were obtained by GPC measurements, are higher than it should be, mainly because the crosslinking reaction between a few of double bonds carried out at the reaction temperature. Mn of APSUA is 7795 and polydispersity index of APSUA is 1.59. The values of molar masses of APSUA are lower than it should be (ideal molar mass is 10,758) and polydispersity index of sample APSUA is higher. It may be attributed to the side reactions. During the synthesis of the adduct, the reaction of isocyanate group with hydroxyl group did not carry out totally in sequence as we expected, and moreover, the side products didn't quite be removed from reaction product. These caused the polydispersity of APUSA.

Compatibility of APSUA with monomers and solubility in dilute alkali solution

APSUA was mixed with monomers HEA, IBOA, DPGDA, TMPTA and 1% sodium carbonate, respectively, with the weight ratio of 20/80, 30/70, 40/60, and 50/50, respectively. The mixtures were stirred for 10 min and stored for 24 h at room temperature prior to a visual observation of the clarity degree

of the mixtures. It was found that APSUA has a good compatibility with the acrylic monomers at the weight ratio investigated, which could enhance the miscibility of the constituents in coatings, and the newly synthesized polymer could dissolve in 1% sodium carbonate solution easily and completely.

Furthermore, DMTA spectra of UV cured films of APSUA (see Fig. 5) show that the systems of APSUA with HEA, IBOA, DPGDA, TMPTA, respectively, had only one glass transition temperature. This further proved good compatibility of APSUA with acrylic monomers.[22] Introduction of chain of urethane and acrylate improves compatibility of APSUA with acrylic monomers.
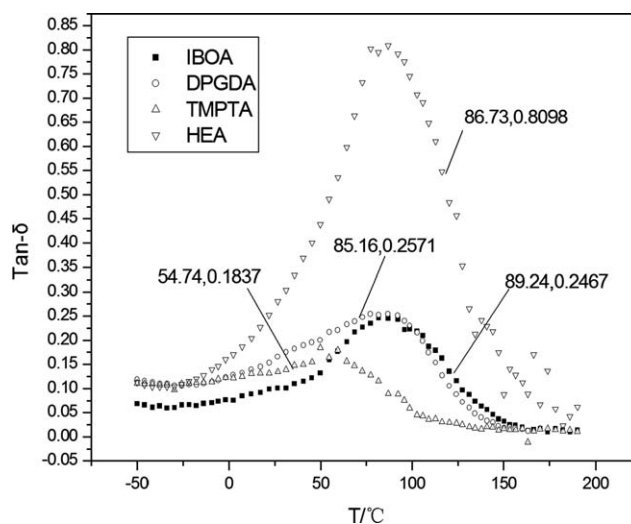

Figure 5 DMTA spectra of the cured films of APSUA systems. Composition of the system: APSUA : monomer : 1173 = 50 : 50 : 0.001 (wt.).
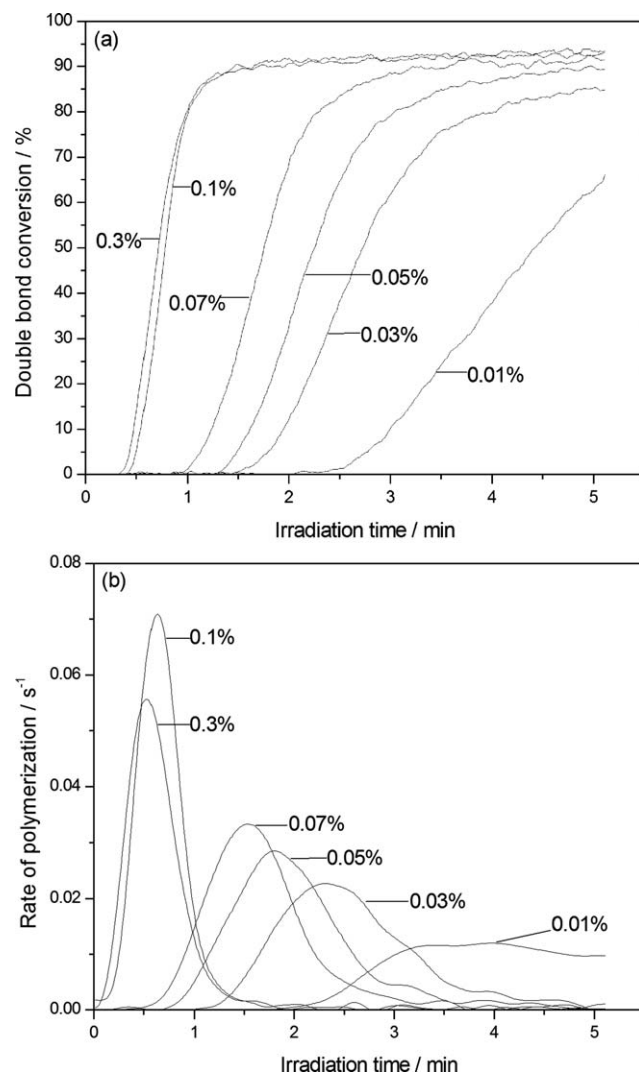
TABLE I
Molecular Weight Distribution of TDDM and APSUA

Oligomers	M_n	M_w	M_P	M_w/M_n
TDDM	983	1216	924	1.24
APSUA	7795	12397	8016	1.59

Figure 6 Effect of concentration of photoinitiator Darocur 1173 on photopolymerization kinetics of the oligomer APSUA. APSUA : DPGDA = 50 : 50 (wt.); (a) the double bond conversion and (b) the rate of photopolymerization as a function of irradiation time.

Photopolymerization kinetics

The most important parameters characterizing the photopolymerization behavior of a multifunctional oligomer are the rate at the peak maximum ($R_p{}^{max}$) and the final degree of double bond conversion (DC) after a given irradiation time.

The rate of photopolymerization is directly related to the number of initiating species generated. And the concentration of photoinitiator is one of the key factors that affected the generation of the initiating species.[23] In this work, the effect of photoinitiator (Darocur 1173) concentration on photopolymerization behavior of APSUA was investigated.

From Figure 6, It is seen that the $R_P{}^{max}$ and double bond conversion both rose with increase of Darocur 1173 concentration, but when photoinitiator concentration increased from 0.01 wt % to 0.3 wt %,

increase of $R_P{}^{max}$ and double bond conversion is not notable. It might be attributed to excessively high concentration of photoinitiator, leading to generation of excess free radicals, which can easily coupled each other to terminate propagation reaction. In addition, when concentration of photoinitiator increased, a large number of initiator molecules on the surface layer absorbed a great amount of light energy, resulting in lower light energy transmitted to deep layer of the system. As a result, conversion of double bond on deep layer decreased. These reasons leaded to that increase of $R_P{}^{max}$ and double bond conversion was not notable, when concentration of photoinitiator was increased to a certain value.

The effect of reactive monomers on photopolymerization kinetics of APSUA is shown in Figure 7.
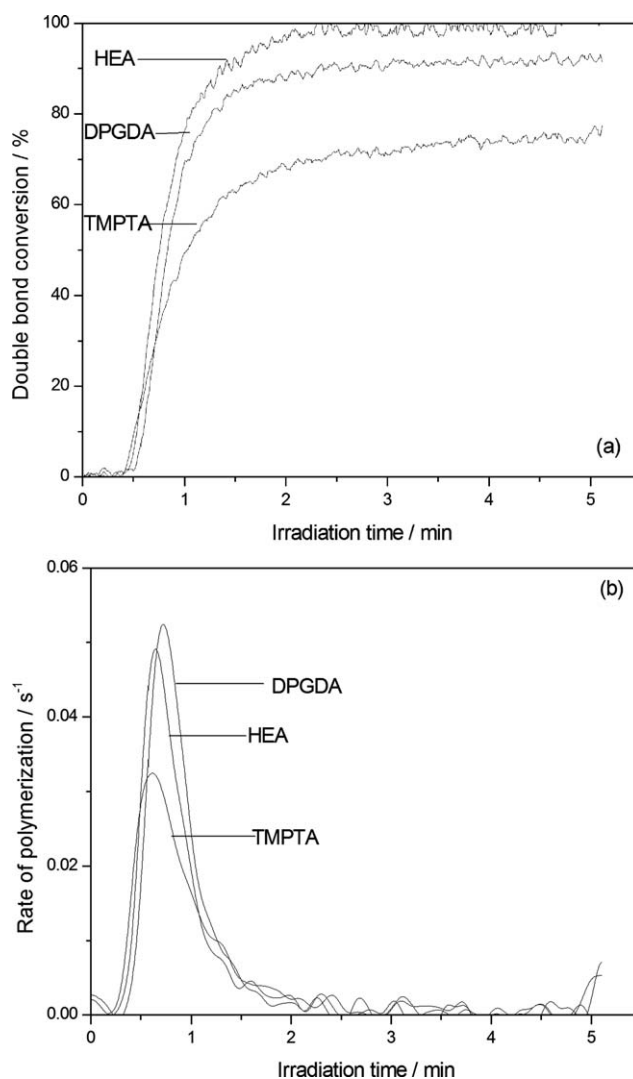

Figure 7 Effect of monomers on photopolymerization kinetics of APSUA. Composition of the system: APSUA : monomer : 1173 = 50 : 50 : 0.001(wt.); (a) the double bond conversion and (b) the rate of photopolymerization as a function of irradiation time.
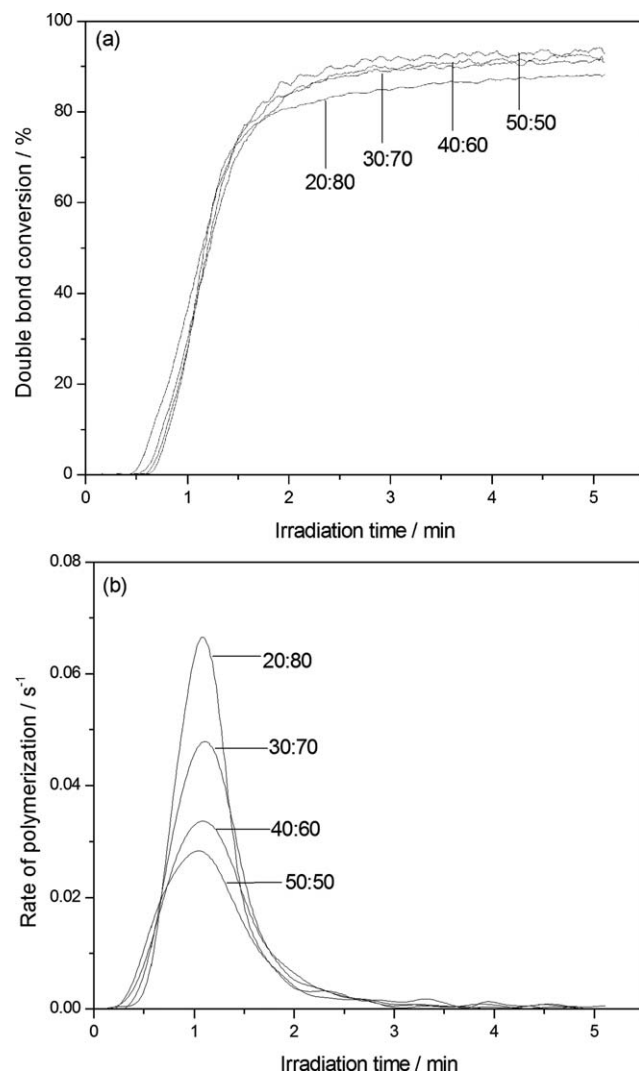
Figure 8 Effect of ratio of APSUA to monomer on photo-polymerization kinetics of APSUA. Composition of the system: (APSUA+DPGDA) : 1173 = 100 : 0.001(wt.); (a) the double bond conversion and (b) the rate of photopolymerization as a function of irradiation time.

R_p^{max} of the systems increases in the following order DPGDA>HEA>TMPTA. It is attributed to that DPGDA has two acrylate groups and lower visco-

Figure 9 Appearance of cured film of APSUA system with image. Composition of the system: APSUA : DPGDA : 1173 = 60 : 40 : 0.0007(wt.). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

TABLE II
Properties of APSUA Cured Film

Items	Value	
	APSUA	OAK-27
Tensile strength (Mpa)	5.60	5.09
Elongation percentage (%)	15.50	9.48
Pencil Hardness	5H	2H
Weight loss (%, 300°C)	6.20	25.76
Water absorption (%, 24 h)	2.40	5.01
Solubility in 1%Na$_2$CO$_3$	excellent	insoluble

Composition of the system: APSUA : DPGDA : 1173 = 50 : 50 : 0.001 (wt.).

sity. HEA contains only one acrylate group, resulting in lower photopolymerization rate of APSUA system with HEA. Although TMPTA contains three acrylate groups, R_p^{max} and double bond conversion of APSUA system with TMPTA is the lowest owing to high viscosity of TMPTA. The double bond conversion of the systems increases in the following order HEA>DPGDA>TMPTA. The reason is that three-dimensional gel structure more easily forms in the system with TMPTA and DPGDA compared with HEA, leading to that uncured double bonds trapped in the polymeric networks cannot polymerize further.[24]

The effect of ratio of APSUA to monomer (DPGDA) on photopolymerization kinetics of APSUA is showed in Figure 8. The change of double bond conversion is not notable, when the ratio of APSUA to DPGDA increased from 20/80 to 50/50, but the R_p^{max} of the systems decreases significantly. It is mainly because of the concentration of acrylate end groups in the system is much lower. Thus, it could more rapidly polymerized under UV-irradiation when the ratio of APSUA to monomer increased from 20/80 to 50/50.

Property of photoimaging

The mixture of APSUA, DPGDA, and 1173 was cured under UV-irradiation through a patterned mask. Then, after development in dilute alkali solution a resist pattern was formed (see Fig. 9). The part which is not cured by ultraviolet light could dissolve easily and completely in 1% sodium carbonate solution. The APSUA is capable of securing accuracy of pattern.

Properties of UV-curing film

The properties of UV-cured film of the APSUA have been measured as those listed in Table II. As shown in Table II, APSUA film possesses both flexibility and toughness, which were proved by the values of hardness (5H), tensile strength (5.6 MPa)

and elongation (15.5%). The thermal weight loss of APSUA (6.20%) is smaller than the commercial flexible polyurethane acrylate OAK-27 (25.76%), which is ascribed to the molecular structure of APSUA. The Si—O bond is one of the most thermally stable bonds which has a higher bond dissociation energy (452 kJ/mol) compared with 359 kJ/mol for C—O, 414.2 kJ/mol for C—C, 347.3 kJ/mol for C—H and 319 kJ/mol for Si—C bond.

CONCLUSIONS

Here we have synthesized a novel environmental friendly alkali-soluble photosensitive polysiloxane urethane acrylate (APSUA). The optimum temperature for the synthesis reaction was found to be 50°C. The structure of the APSUA was characterized by ^{1}HNMR, IR, and GPC. APSUA is excellent in compatibility with most of acrylate monomers. The photopolymerization properties of system of APSUA with all kinds of monomers were investigated by RT-IR. The polymerization rate of APSUA system increased gradually with increase of concentration of Darocur 1173 and the optimum concentration of photoinitiator was 0.1wt %. The functionality, concentration and viscosity of the monomer have great influence on the polymerization kinetics of APSUA system. The system of APSUA with DPGDA monomer has higher polymerization rate (0.0521 s^{-1}) and double bond conversion (93.2%). The APSUA system with acrylate monomers could form regular image under UV-irradiation through a patterned mask. The cured coating film of the APSUA system possesses excellent flexibility, toughness, and heat resistance. APSUA not only maintains the properties of polysiloxane, but also could rapidly polymerize under UV-irradiation. These properties offer APSUA potential for applications in solder resist ink. Especially, the APSUA system could dissolve in 1% sodium carbonate solution easily and completely, which could be more energy-saving and environmental friendly in industrial production.

References

1. Tsai M. F.; Lee Y. D.; Long Y. C. J Polym Res 2000, 7, 73.
2. Bauer F.; Gläsel, H. J.; Decke, U. Prog Org Coat 2003, 47, 147.
3. Sung, P. H.; Lin, C. Y. Eur Polym Mater 1997, 33, 231.
4. Bartlett, I. D.; Marshall, J. M.; Maud, J. M. J Non-Cryst Solids 1996, 198, 665.
5. Vlastimil, M.; Klaus, R.; Miloš, H.; Marie, P. Sens Actuators B Chem 1997, 39, 438.
6. Li, L. H.; Xu, W. J.; Wu, Y. M.; et al. Prog Org Coat 2005, 53, 77.
7. Kayaman-Apohan, N.; Amanoel, A.; Arsu, N.; Gungor, A. Prog Org Coat 2004, 49, 23.
8. Samuelsson, J.; Sundell, P. E.; Johansson, M. Prog Org Coat 2004, 50, 193.
9. Bongiovanni, R.; Sangermano, M.; Malucelli, G.; Priola, A. Prog Org Coat 2003, 53, 46.
10. Kim, H. K.; Hong, J. W. Eur Polym Mater 2003, 39, 2235.
11. Wang, W. Z.; Cheng, K. X. Eur Polym Mater 2003, 139, 1891.
12. Choi, K. H.; Jung, J C. Polym Adv Technol 2005, 16, 387.
13. Baltazar, E. H.; Reyes-Sandoval, G. A. Col Surf A: Phys Eng Aspects 2004, 238, 13.
14. Seng, W. P.; Tam, K. C. Col Surf A: Phys Eng Aspects 1999, 154, 365.
15. Tan, H.; Tam, K. C. J Non-Newtonian Fluid Mech 2000, 92, 167.
16. Qing, F. S.; Xiao, D. F.; Liu, Y. Y.; Kong, J. J. Polym Sci Part A-Polym Chem 2006, 44, 3261.
17. Schonhorn, H.; Torza, S.; Albarino, R. V.; Vazirani, H. N.; Wang, T. T. J Appl Polym Sci 1979, 23, 75.
18. Bong, T. H.; Kwon, S. S.; Dae, S. K. J Appl Polym Sci 2005, 98, 1180.
19. Xiong, J.; Sun, F.; Du, H. G.; Chin, J. Analysis Laboratory 2007, 26, 73.
20. Scherzer, T.; Mehnert, R.; Volland, A.; Lucht, H. Nucl Instrum Methods Phys Res Sect B 2005, 236, 123.
21. Li, S. J.; He, Y.; Nie, J. J. Photochem Photobiol A: Chem 2007, 191, 25.
22. Sun, F.; Shi, J. Prog Org Coat 2009, 66, 412.
23. Sun, F.; Jiang, S. L.; Liu, J. Nucl Instrum Methods Phys Res Sect B 2007, 264, 318.
24. Huang, Z. G.; Shi, W. F. Prog Org Coat 2007, 59, 312.