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Synthesis and UV-Curing Behaviors of Urethane Acrylic Oligomers Modified by the Incorporation of Silicone Diols into the Soft Segments for a 3D Multi-Chip Package Process

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Ultraviolet (UV)-curable urethane acrylic oligomers were synthesized and then modified by the incorporation of silicone diols with different molecular weights to improve thermal stability for temporary bonding and debonding adhesives in a three-dimensional multi-chip package process. The UV-curing behaviors were investigated using photo-differential scanning calorimetry, Fourier transform infrared spectroscopy-attenuated total reflectance, and gel fraction, while the UV-curing kinetics was also studied. In addition, the thermal stability of the samples was checked using thermogravimetric analysis. UV-curing and thermal stability were more affected by the molecular weight of the silicone diols than by UV dose due to both the flexibility and the steric hindrance of the synthesized oligomer structures.

Key words: Temporary bonding and debonding adhesives, urethane acrylic oligomers, silicone diols, curing behaviors, 3D multi-chip packages process

INTRODUCTION

Integration of multi-media functions using three-dimensional (3D) integrated circuit (IC) packaging technology has become increasingly essential and crucial due to the ever-increasing demands for mobile devices with great functional diversification, heterogeneity, miniaturization and high performance. To meet the growing challenges, in addition to the revolution of IC circuit design, great engineering efforts are presently being made in the development of high-reliability, high-performance 3D chip stacking packaging with micro-bump and through-silicon via (TSV) technology.[1] As compared to other flip chip technology,[2–4] there are many implied advantageous features in the packaging technology, including increased function density, reduced package profile and interconnect length, and enhanced electrical performance. So, considerable 3D IC packaging technologies have been proposed.[5–10]

In the most commonly used adhesive wafer bonding processes, a polymer adhesive is applied to one or both of the wafer surfaces to be bonded. After joining the wafer surfaces that are covered with the polymer adhesive, pressure is applied to force the wafer surfaces into intimate contact. The polymer adhesive is then converted from a liquid or viscoelastic state into a solid state, typically done by exposing the polymer adhesive to heat or ultraviolet (UV) light. The main advantages of adhesive wafer bonding include relatively low bonding temperatures, between room temperature and 450°C depending on the polymer material, insensitivity to the topology of the wafer surfaces, compatibility with standard complementary metal-oxide semiconductor (CMOS) wafers and the ability to join practically any wafer materials. Adhesive wafer bonding does not require special wafer surface treatments such as planarization or excessive cleaning. Structures and particles at the wafer surfaces can be tolerated and offset to some extent by the polymer adhesive. While adhesive wafer

Synthesis and UV-Curing Behaviors of Urethane Acrylic Oligomers Modified by the
Incorporation of Silicone Diols into the Soft Segments for a 3D Multi-chip Package Process

2407

bonding is a comparably simple, robust, and low-cost process, concerns such as the limited temperature stability and limited data about the long-term stability of many polymer adhesives in demanding environments need to be considered. Also, adhesive wafer bonding does not provide hermetically sealed bonds towards gasses and moisture.[11,12] The process for temporary wafer bonding is shown in Fig. 1[13] and is mentioned in previous studies.[14–17]

Silicones have become important commercial polymers because of a combination of properties, including high thermal stability,[18] low surface tension, and low glass transition temperature.[18,19] However, the materials have relatively poor mechanical strength,[20] which is usually attributed to flaws or microcracks that readily grow because of the high mobility of the chains.[21] In order to improve the mechanical properties of the silicone materials while retaining the inherent properties of silicone moiety, numerous synthetic pathways have been attempted. The methods adopted are usually blending of siloxanes with hydrocarbon-based polymers.[22,23] But the non-polar nature of the siloxanes together with the very low levels of intermolecular attractions lead to thermodynamically and mechanically incompatible blends with virtually all other polymeric systems. This is reflected in the very low solubility parameter of siloxanes when compared to other polymers.[24] Therefore, it is more useful to synthesize a polymer that possesses both the polysiloxane and other hydrocarbon-based polymers. Thus, polydimethylsiloxane (PDMS) has been introduced to various polymeric systems, such as polystyrene,[25] poly(methacrylate),[26] polycarbonate[27] and polyurethanes[28,29] through copolymerization methods.

Photo-induced reactions comprise polymerization introduced by UV light, visible light, electron beam (EB) or laser. Photopolymerization science and technology has attracted a significant amount of attention due to its various industrial applications, such as inks, coatings, photoresists and adhesives.[30,31] Among the various methods of photo-curing, UV-curing systems are widely used due to advantages such as their rapid production rate in a small place of work, lower process costs, high chemical stability, high dimensional stability, high thermal stability and solvent-free curing at ambient temperatures.[32,33]

The aim of this study is twofold. Firstly, synthesis of silicone-modified urethane acrylic oligomers for a 3D multi-chip package process, in which the synthesized oligomers have the incorporation of different chain length of silicone diols in the soft segments whose pathways are investigated by Fourier transform infrared (FTIR) spectroscopy and ^{1}H nuclear magnetic resonance (^{1}H NMR) analysis. The second goal is to carry out UV-curing behaviors by photo-DSC, gel fraction, and TGA depending on different chain lengths of the Si–O groups. This study will provide a fundamental direction in the design of temporary bonding and debonding adhesives for 3D multichip packaging processes since the thermal stability and UV-curing behaviors can be adjusted by the content of Si–O in the synthesized oligomers.

EXPERIMENTAL

Materials

Table I shows the chemical structures and properties of the raw materials used for the synthesis process. Carbinol polysiloxane diol (Shin-Etsu, Japan) and isophorone diisocyanate (IPDI; Bayer Material Science) were dried at 100°C. Hydroxyethyl methacrylate was used as end-capping materials without pretreatment. Approximately 0.1 wt.% of dibutyltin dilaurate was added to cause the reaction to take place at a rapid rate as a catalyst in a urethane reaction. Hydroxydimethyl acetophenone (Micure HP-8; Miwon Specialty Chemical, Korea) was used as the photo-initiator for UV-curing, as shown in Fig. 2.[34]

Synthesis of Silicone-Modified Urethane Acrylic Oligomers

Figure 3 shows the synthesis process scheme of the UV-curable silicone-modified urethane acrylic oligomers. The reaction time was determined by observing changes in the FT-IR peak at 2265 cm^{-1}
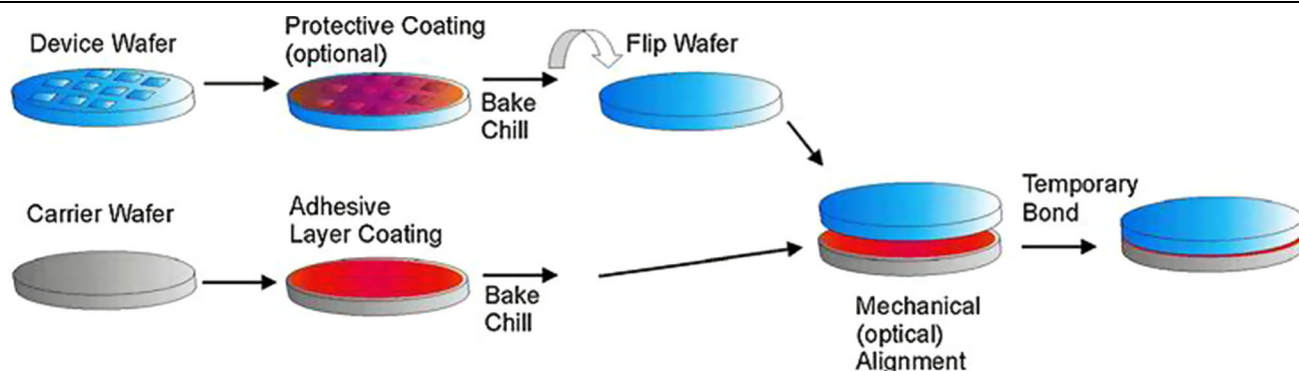

Fig. 1. The process for temporary wafer bonding.[13]

Table I. Raw materials used for urethane acrylic oligomers modified by the incorporation of silicone diols

Function	Materials	Abbreviation	Chemical structure	Molecular weight ($g\ mol^{-1}$)	Supplier
Polyol	Carbinol polysiloxane diol	–		940/1810/3206/5100	Shin-Etsu
Isocyanate	Isophorone diisocyanate	IPDI		228	Bayer Material Science
End-capping agent	Hydroxyethyl methacrylate	HEMA		130	Sigma Aldrich

Each sample has different M_w ($g\ mol^{-1}$) of carbinol polysiloxane diols as follows: PSD-I (hydrocarbon diols not polysiloxane diol); PSD-II (940 $g\ mol^{-1}$ of polysiloxane diol); PSD-III (1810 $g\ mol^{-1}$ of polysiloxane diol); PSD-IV (3206 $g\ mol^{-1}$ of polysiloxane diol); PSD-IV (5100 $g\ mol^{-1}$ of polysiloxane diol).
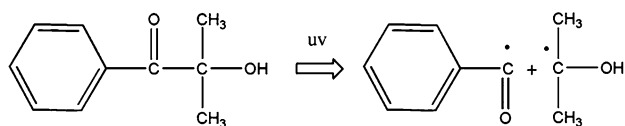

Fig. 2. Cleavage of hydroxydimethyl acetophenone as a photo-initiator in the presence of UV light.[34]

(NCO peak), which decreased with the polyurethane reaction. Initially, IPDI was charged into a dried 300-mL round-bottomed flask equipped with a four-necked separable flask with a mechanical stirrer, thermometer and condenser with a drying tube and an N_2 inlet. The temperature was maintained at room temperature with constant stirring. The four types of hydroxy-terminated carbinol polysiloxanes which have different molecular weight with some of the catalyst (dibutyltin dilaurate) was then added dropwise over a period of 5 h and was maintained for a further 1 h. The reaction temperature was increased to 50°C using a constant temperature heating mantle with constant stirring. Hydroxyethyl methacrylate was added dropwise over a period 1 h and was reacted for 3 h until the NCO peak had almost disappeared. After cooling to ambient temperature, a photo-initiator was blended to initiate UV-curing.

UV-Curing Behaviors

Photo Differential Scanning Calorimetry (Photo-DSC)

The photo-DSC experiments were carried out using a DSC (Q-1000, TA Instruments, USA) equipped with a photocalorimetric accessory (Novacure 2100), which had light from a 100-W medium-pressure mercury lamp (main wave length: 250–650 nm) to investigate the UV-curing behaviors. The light intensity was employed by placing an empty DSC pan on the sample cell. The UV light intensity at the sample was 50 mW cm^{-2}.

The weight of the sample was approximately 2 mg and the sample was placed in an open aluminum DSC pan. Before UV-curing, the samples were evaporated at 50°C for 10 m. The measurements were carried out at 30°C in flowing N_2 gas at 50 mL m^{-1}.

Fourier Transform Infrared Spectroscopy (FT-IR)

The IR spectra were obtained using a JASCO FTIR-6100 (Japan) equipped with an attenuated total reflectance (ATR) accessory. In order to obtain the IR spectra of dual-curable adhesives, the cured adhesive samples were cut into $0.5 \times 0.5\ cm^2$ pieces. The ATR prism was a diamond and its refractive index at 1000 cm^{-1} was 2.4 with a transmission range from 4000 cm^{-1} to 650 cm^{-1}. The resolution of the spectra recorded was 4 cm^{-1}. The curing behavior of the UV-curable oligomers was analyzed by observing the changes in the C=C bonding bands. All FTIR-ATR spectra were modified by baseline correction. The conversion (%) was calculated by the equation[35]

$$\text{Conversion }(\%) = \left[(A_{810}/A_{1730})_0 - (A_{810}/A_{1730})_t \right] / (A_{810}/A_{1730})_0 \times 100 \tag{1}$$

In this equation, $(A_{810}/A_{1730})_0$ and $(A_{810}/A_{1730})_t$ denote the relative absorbance of the C=C bonds before curing and at a given curing time t, respectively.

Gel Fraction

Gel fraction determination is a convenient method of measuring insoluble fractions, such as the fractions of cross-linked or network polymers. The gel fraction is the insoluble part of the polymer in the solvent. It is a significant factor when investigating curing behaviors. The gel fraction of the

Synthesis and UV-Curing Behaviors of Urethane Acrylic Oligomers Modified by the
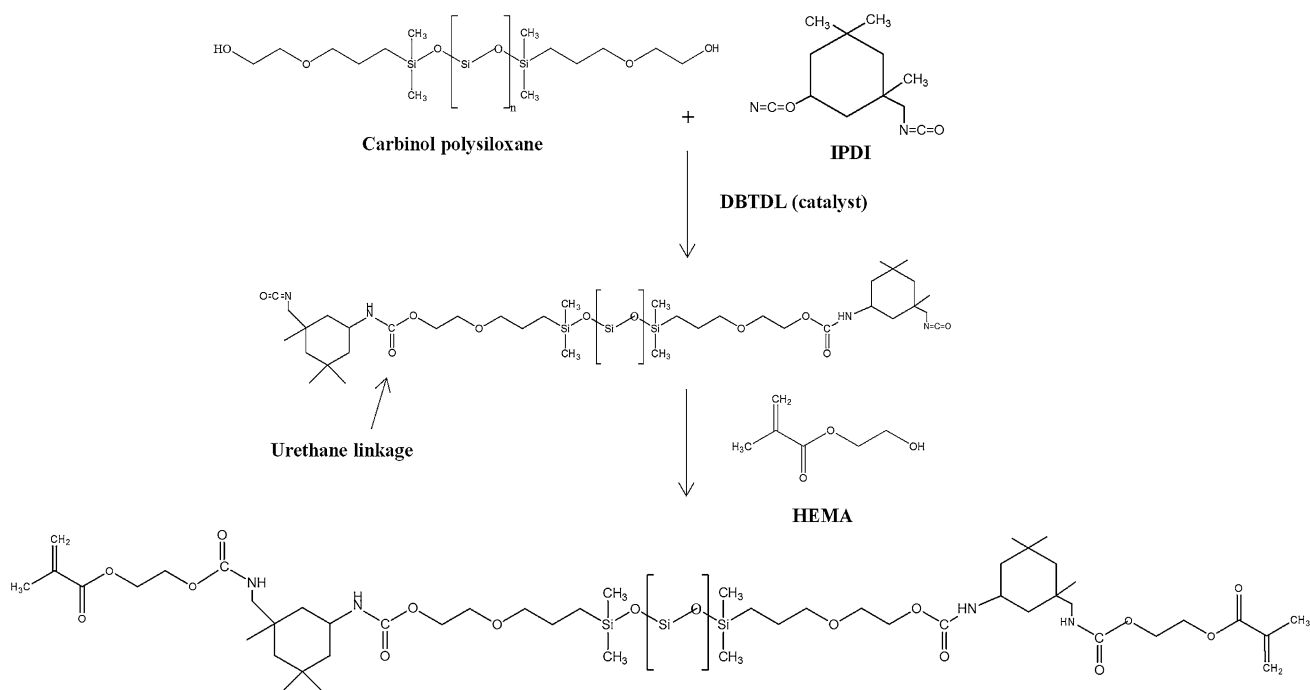Incorporation of Silicone Diols into the Soft Segments for a 3D Multi-chip Package Process

2409

Fig. 3. The synthesis process scheme of the UV-curable silicone-modified urethane acrylic oligomers.

UV-curable oligomers after UV-curing was determined by soaking it in toluene at 50°C for 24 h. The insoluble part was removed by filtration and dried at 50°C to a constant weight. The gel fraction was calculated by applying the following equation:

$$\text{Gel fraction} = (W_1 / W_o) \quad (2)$$

Here, W_o is the initial weight of the sample and W_1 is the weight of the dry. The test was replicated ten times.

Thermogravimetric Analysis (TGA)

The thermal stability and decomposition profiles of the dual-curable adhesives were measured using a thermogravimetric analyzer (PerkinElmer Thermogravimetric Pyris 1 TGA model). The samples were loaded into a ceramic pan, and heated from 25°C to 200°C at a constant heating rate of 10°C m^{-1} in an inert nitrogen atmosphere.

RESULTS AND DISCUSSION

UV-Curing Behaviors

Photo Differential Scanning Calorimetry (photo-DSC)

The kinetics of the UV-curing is very important to study for employing UV technology to each application area. However, monitoring the UV-curing behavior is very difficult because of its extremely rapid speed.[36] So, photo-DSC was employed for the

UV-curing behavior owing to its method for evaluating the reactivity and measuring the kinetics of the reaction. Figure 4 presents the isothermal UV-curing heat enthalpy and conversion profiles of the samples. The heat flow (W g^{-1}) plotted in Fig. 4a was measured by photo-DSC and other plots (Fig. 4b–d) were explained by further calculations. The reaction enthalpy was calculated by integrating the area under the exothermic peak using the following equation[37,38]:

$$\alpha = \Delta H_t / \Delta H_o^{\text{theor}} \quad (3)$$

where ΔH_t is the reaction heat enthalpy released at time t and ΔH_o^{theor} is the theoretical heat enthalpy for complete conversion. $\Delta H_o^{\text{theor}}$ of methacrylate is 54.4 kJ mol^{-1} (13.1 kcal mol^{-1}).[39,40] The heat of UV-induced polymerization of each sample ($\Delta H_o^{\text{theor}}$ (*sample*) was calculated using the following equation:

$$\Delta H_o^{\text{theor}}(\text{sample}) = \Delta H_o^{\text{theor}}(\text{methacrylate}) / \\ M_w^{\text{theor}} \times \text{Functionality} \quad (4)$$

where M_w^{theor} is the theoretical molecular weight of the repeating unit. Table II shows the calculated results of all the samples. The rate of polymerization (R_p) is related directly to heat flow (dH/dt) using the following equation[37,38];

$$R_p = \text{d}\alpha/\text{d}t = (\text{d}H/\text{d}t) / \Delta H_o^{\text{theor}} \quad (5)$$

where dα/dt is the conversion rate or the rate of polymerization, $\Delta H_o^{\text{theor}}$ is the total exothermic heat
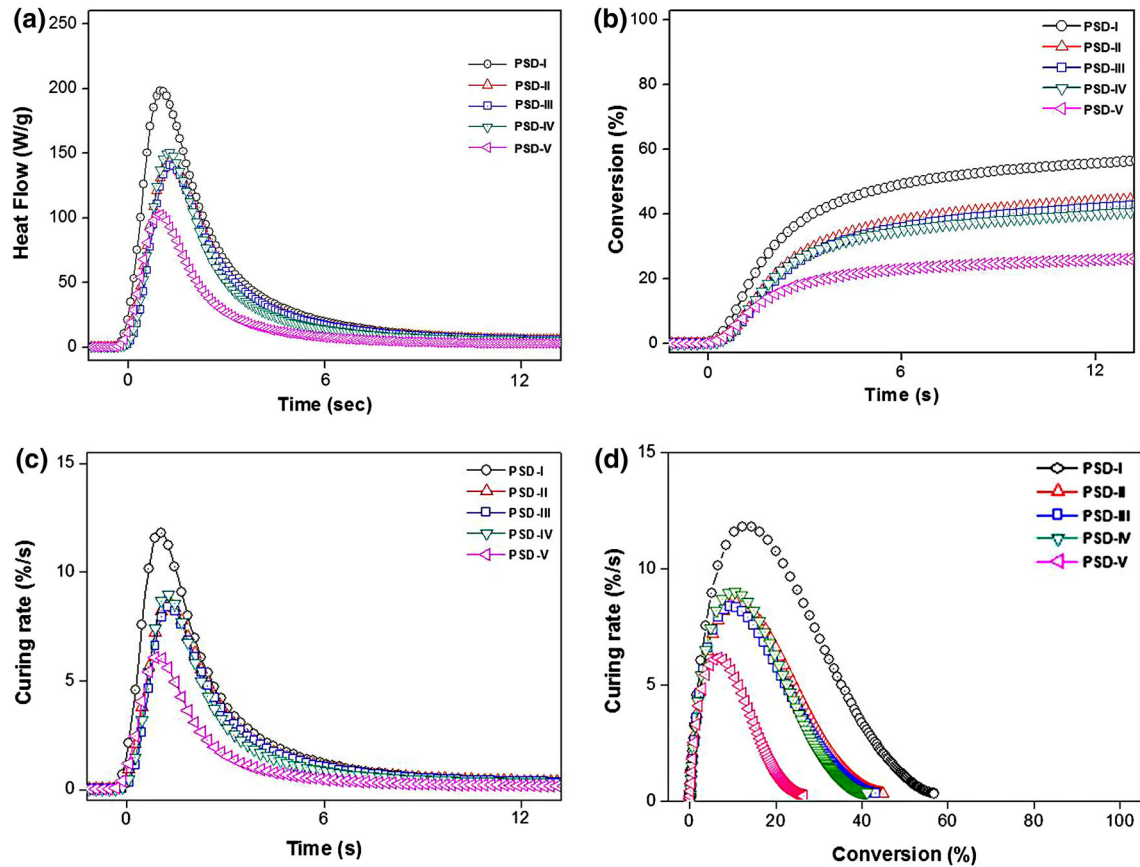
Fig. 4. Isothermal UV-curing heat enthalpy and conversion profiles of the samples by photo-DSC: (a) heat flow versus time, (b) conversion versus time, (c) conversion rate versus time and (d) conversion rate versus conversion.

Table II. Calculated theoretical reaction enthalpy of UV-curable urethane acrylic oligomers modified by the incorporation of silicone diols

Samples	Theoritical M_w of repeating unit	Functionality (methacrylate)	Theoretical reaction enthalpy			
			kJ mol^{-1}	J g^{-1}	Kcal mol^{-1}	Cal g^{-1}
PSD-I	1480	2	110.0	74.4	26.2	17.7
PSD-II	1656	2	109.6	66.4	26.1	15.8
PSD-III	2526	2	110.0	43.7	26.2	10.4
PSD-IV	3922	2	110.0	28.1	26.2	6.7
PSD-V	5816	2	109.6	18.9	26.1	4.5

of reaction and dH/dt is the measured heat flow at a constant temperature.

In a previous study, UV-curing behaviors were affected by several factors such as UV intensity, UV dose, species, content of photoinitiator, reaction temperature, and the reactivity of functional groups.[37] Figure 4a presents the exothermic heat flows. At the beginning of the reaction, the early onset of auto-acceleration by the activation of radicals occurred as a steep increase, which was followed by auto-deceleration as indicated in the rapid dropping curves.[36] A minor difference was shown

among the samples with different molecular weight of silicone diols. The curing rate and conversion were decreased with increasing molecular weight of silicone diols because of relatively low reactivity of functional groups. The conversion of each sample was calculated using Eqs. 3 and 4 in Fig. 4b. The final conversion was decreased with the increasing molecular weight of silicone diols. Also, Fig. 4d shows the plots of the curing rate versus conversion of the samples. In the initial slope, the curing rates were increased by increasing the conversion regardless of the different molecular weight of the

Synthesis and UV-Curing Behaviors of Urethane Acrylic Oligomers Modified by the
Incorporation of Silicone Diols into the Soft Segments for a 3D Multi-chip Package Process
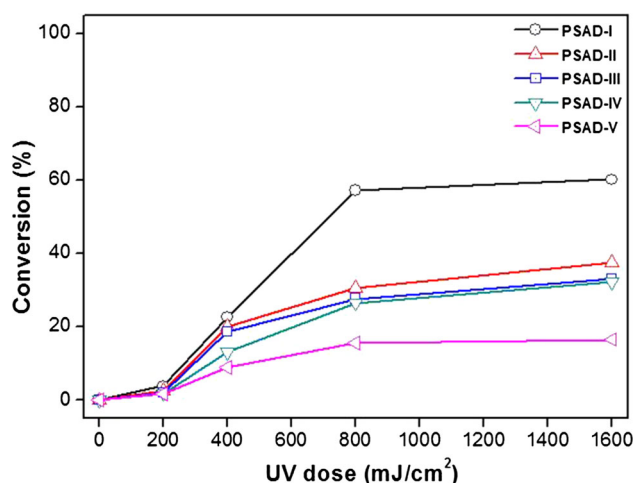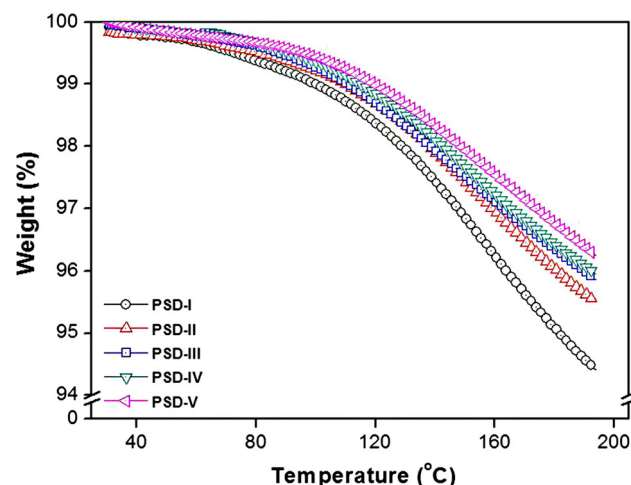
2411

Fig. 5. C=C bonds conversion with increasing UV dose by FT-IR.

Fig. 7. Thermogravimetric analysis (TGA) of UV-cured samples.

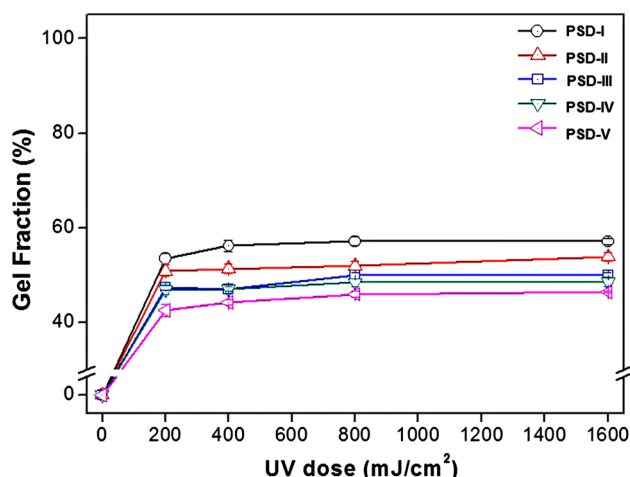Fig. 6. Gel fraction with increasing UV dose.

acceleration). In the propagation step, polymerization proceeds quite rapidly at first with high double bond conversion due to a vitrification effect caused by the UV-cured glassy polymer networks (auto-deceleration).[42–45] Figure 5 indicates the effect of the different molecular weights of the silicone diols on the C=C dond conversion during the UV-curing process. In the plot, the plateau value at the end of curing means the final conversion while the initial slope of the conversion is the UV-curing rate. The curing rate and final conversion were affected by different molecular weights of silicone diols which means that the sample with the lower molecular weights of silicone diols has more reactivity than any other sample due to the flexibility and the lower steric hindrance. This result was similar to that of the C=C bond conversion measured by photo-DSC.

Gel Fraction

Figure 6 shows the relationship between the UV dose and the gel fraction of the samples with different molecular weights of the silicone diols. As the UV dose increases, the concentration of free radicals increases, causing the crosslinking density to increase due to a three-dimensional crosslinked insoluble and infusible network. In all samples, the gel fraction was constant regardless of molecular weights of silicone diols at higher than 800 mJ cm^{-2} of UV dose. But the gel fraction increased with an increase in the UV dose, respectively. As mentioned before, in the sample with higher molecular weight of silicone diols, the gel content conspicuously decreased, because of the low reactivity regardless of UV dose.[46] In high UV doses of the samples, the radical concentration is high and more chain scission reactions are induced, continuing for some time after UV irradiation.[47] Considering that for higher radiation doses scission reactions are supposed to compete with crosslinking, the increase in the

silicone diols. But they were decreased at over 20% of the conversion. Among all the samples, the sample with the lowest molecular weight of silicone diols had the highest curing rate in all conversions, which means the curing rate and conversion is influenced by the chain of flexibility and the steric hindrance depending on the molecular weight of silicone diols.

FTIR-ATR Spectroscopy

The UV-curing rate and final C=C bond conversion depend strongly on the functionality and chain mobility. They also have a close correlation with the crosslink density and affect the physical properties of UV-cured film.[41,42]

Generally, the chemistry of radical initiated photo-polymerization can be divided into the initiation, propagation and termination. In the initiation step, a photoinitiator is excited and produces a radical species as a result of the UV energy, which can initiate radical polymerization (auto-

extent of crosslinking can be accounted for by the fact that chain scissions lead to entanglement couplings which act as crosslinks.[48]

Thermogravimetric Analysis (TGA)

An investigation of the thermal degradation process provides methods for the determining optimum conditions for manipulating and processing the polymer as well as obtaining high performance polymers with improved thermal stability.[49] Also, accelerating the lifetime testing of polymers can be possible by short-term experiments used to predict the in-use lifetime through thermogravimetric analysis.[50] Figure 7 shows the thermal stability of samples at temperatures ranging from 25°C to 200°C. It indicates the weight loss curves and derivatives of the weight profile of samples, respectively. In a previous study, the UV-cured samples without silicone diols were decomposed in two steps. The first part of degradation correlates with the hard segment (IPDI–HEMA segment) of the polyurethane chain, and the second peak shows degradation of the soft segment (polycarbonate doil).[51,52] However, the UV-cured samples with silicone diols had three decomposition peaks. The first and second peaks were for the hard and soft segments, respectively, as mentioned above. The third peak reflects the decomposition of the silicone diols. Incorporation of silicone diols increased the thermal stability of the polyurethane (meth)acrylate due to the greater thermal stability of the silicone diols component.[53] Hence, the thermal stability of the samples is more affected by the molecular weight of silicone diols rather than gel fraction investigated by the crosslinking density.

CONCLUSION

UV-curable urethane acrylic oligomers were modified by the incorporation of the silicone diols into the soft segments to improve the thermal stability for temporary bonding and debonding adhesives in 3D multichip packaging process. The effect of the molecular weight of the silicone diols on the properties was studied by end-capping with hydroxymethacrylate. The UV-curing behaviors investigated by photo-DSC, FTIR-ATR, and gel fraction were strongly dependemt on the molecular weight of the silicone diols. Also, the thermal stability was increased by increasing the molecular weight of the silicone diols. Based on the above analysis, the results are going to be fundamental and meaningful data in designing thermally stable temporary bonding and debonding adhesives.

ACKNOWLEDGEMENTS

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