# Photoacid-generating Sulfonyloxymaleimide Polymers and Their Application to Photoimaging

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SUMMARY: Synthesis and properties of photoacid generating polymers based on four sulfonyloxymaleimides (RsOMI), *N*-(tosyloxy)maleimide (TsOMI), *N*-(methanesulfonyloxy)maleimide (MsOMI), *N*-(trifluoromethanesulfonyloxy)maleimide (TfOMI), and *N*-(10-camphorsulfonyloxy)maleimide (CsOMI) are described. The RsOMI polymers photochemically produce corresponding sulfonic acids (RsOH), TsOH, MsOH, TfOH and 10-camphorsulfonic acid (CSA) which can catalyze deprotection of the acid-labile t-BOC groups in the same polymer chains via a chemical amplification process. The photoacid generating polymers with concurrent acid-labile groups offer great potential for applications in functional transformation and photoimaging.

## Introduction

Great deals of research efforts have been made for advancement of *N*-substituted functional maleimide monomers ($R_fMI$) and their polymers[1]. Functional maleimides offer a few unique features as follows: facile synthesis by introduction of a desired functionality into the imino nitrogen, relatively low reactivity in radical homopolymerization so as to handle and store without special cautions, and sufficiently high reactivity in copolymerization by electron-donor/acceptor (EDA) systems so as to obtain high molecular weight polymers with high conversions. Previously, we have reported on the *N*-substituted functional polymaleimides having acid-labile protecting groups represented by the *tert*-butyloxycarbonyl (t-BOC) group[2]. Those maleimide polymers have been applied to microlithography as chemical amplification resists in a deep UV (DUV) region. Polymaleimides containing trialkylsilyl moieties have been also studied as both the acid-labile polymers and photoresist materials[3,4].

In designing an advanced photoresist system based on the chemical amplification concept, it has to incorporate two basic components, namely, an acid-labile polymer and a photoacid generator (PAG) which should generate a strong acid by UV irradiation[5,6]. There has been reported numerous photoacid generating systems from the well-known onium salt PAGs to pure organic PAGs[7]. Nonionic sufonate PAGs have been extensively studied because they offer the advantages of better solubility and being free of metal ions compared to onium salt

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PAGs[8,9]. Among them, 2-nitrobenzyl sulfonates have received special attention as a useful PAG because of ease of synthesis, good solubility, and low UV absorption[8-11]. Most of the reported organic PAGs were low-molecular-weight compounds exemplified by 2-nitrobenzyl sulfonates and *N*-(tosyloxy)phthalimide which generate sulfonic acids by UV irradiation and they have been applied in photoimaging as efficient PAGs[9,12,13]. Polymeric PAGs are considered to be advantageous in terms of thermal stability and compatibility or mixing in the polymer matrix as well as nonvolatility in comparison with the corresponding small molecule PAGs. Only a few polymeric PAGs have been reported, including copolymers of 2-nitrobenzyl *p*-styrenesulfonate[14] and 9-fluorenylideneimino *p*-styrenesulfonate[15] which produce polymer-bound sulfonic acids upon DUV exposure. The copolymers having *p*-(*tert*-butyloxycarbonyloxy)styrene (t-BOCSt) were utilized as a single-component resist system without any externally added PAG.

In this regard, synthesis and properties of the photoacid generating polymers based on four kinds of sulfonyloxymaleimides (RsOMI), *i.e.*, *N*-(tosyloxy)maleimide (TsOMI), *N*-(methanesulfonyloxy)maleimide (MsOMI), *N*-(trifluoromethanesulfonyloxy)maleimide (TfOMI), and *N*-(camphorsulfonyloxy)maleimide (CsOMI) are described[16-20]. The RsOMI units in the polymers were designed to be the potential PAGs because phthalimido-sulfonates were utilized to generate sulfonic acids when irradiated with UV light[9,13]. In particular, the CsOMI polymers considered to have desired properties due to the bulky camphorsulfonate group both PAG itself and subsequently generated acid. The capability of photochemical acid generation and applicability of the RsOMI polymers as a single-component resist system were investigated.

## Results and Discussion

### Synthesis and Polymerization of Sulfonyloxymaleimides[16,18,20]

Four sulfonyloxymaleimides (RsOMI), TsOMI, MsOMI, TfOMI, and CsOMI have been prepared by a retro-Diels-Alder reaction through thermolysis of the corresponding RsOMI/furan adducts as described in Scheme 1. The synthetic results of the adducts and RsOMIs are listed in Table 1. The RsOMI monomers as an electron-acceptor possess high radical polymerizability in EDA copolymerization with electron-donating styrene monomers (XSt) such as styrene (St), *p*-methylstyrene (MeSt), *p*-hydroxystyrene (HOSt), *p*-acetoxy-

Scheme 1

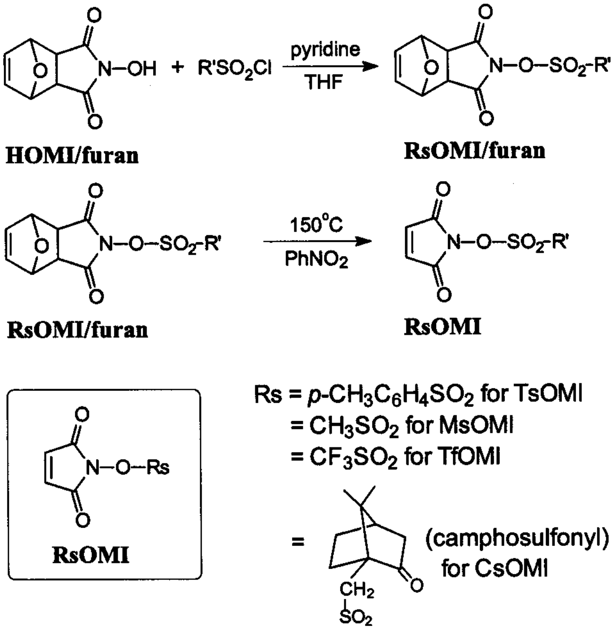
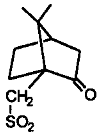


Table 1. Synthetic results of RsOMI/furan adducts and RsOMIs

| Rs | RsOMI/furan | | RsOMI | | RsOMI |
|---|---|---|---|---|---|
| | mp (°C) | yield (%) | mp (°C) | Yield (%) | |
| $CH_3C_6H_4SO_2$ | 165 | 85 | 150 | 57 | TsOMI |
| $CF_3SO_2$ | 116 | 91 | 56 | 75 | TfOMI |
| $CH_3SO_2$ | 165 | 92 | 113 | 65 | MsOMI |
|  | 139 | 89 | 102 | 70 | (+)CsOMI |
| | 143 | 90 | 112 | 70 | (±)CsOMI |

styrene (AcOSt), and t-BOCSt. Terpolymers based on RsOMI and t-BOCSt were also prepared by use of maleimide (MI), *N*-(hydroxy)maleimide (HOMI), *N*-(*p*-hydroxyphenyl)-maleimide (HOPMI), or *N*-(*tert*-butyloxycarbonyloxy)maleimide (t-BOCMI) as the third maleimide monomer (YMI). The unsubstituted (unprotected) MI, HOMI, and HOPMI units were introduced into the terpolymers to improve photosensitivity and adhesion to substrates since their polymers are regarded as "partially deprotected" forms. The copolymers and terpolymers derived by EDA polymerizations of RsOMIs are shown in Scheme 2.

Copolymerizations of RsOMIs with styrene monomers were carried out with 1 to 1 molar feed ratio, and in terpolymerizations, RsOMI, YMI as maleimide (MI, HOMI, HOPMI, or t-BOCMI), and t-BOCSt were fed in the molar ratio of 1:1:2. The results of the radical copolymerizations were summarized in Table 2. The polymers were obtained in high yields over 85% in relatively short time usually within 3 hours. The polymers have inherent viscosity in the range of about 0.5 to 1.0 dL/g in dioxane (Table2). The molecular weights of

Table 2. Radical polymerizations of RsOMIs with styrenes (XSt)

| Polymer[a] | AIBN[b] (mol%) | M/S[c] (g/ml) | Time (h) | Yield (%) | Inherent viscosity[d] |
|---|---|---|---|---|---|
| P(CsOMI/St) | 2 | 0.54 | 3 | 88 | 0.73[e] |
| P(CsOMI/HOSt) | 2 | 0.35 | 5 | 65 | 0.43 |
| P(CsOMI/t-BOCSt) | 2 | 0.55 | 2 | 89 | 0.73[f] |
| P(CsOMI/MI/HOSt) | 2 | 0.50 | 5 | 84 | 0.70 |
| P(TsOMI/St) | 1 | 0.25 | 3 | 91 | 0.55 |
| P(TsOMI/t-BOCSt) | 1 | 0.50 | 2 | 86 | 0.81 |
| P(TsOMI/MI/t-BOCSt) | 1 | 0.40 | 3 | 93 | 0.94 |
| P(TsOMI/t-BOCMI/t-BOCSt) | 0.5 | 0.33 | 5 | 91 | 0.69 |
| P(TfOMI/St) | 1 | 0.33 | 2 | 80 | 0.90 |
| P(TfOMI/t-BOCSt) | 1 | 0.33 | 3 | 87 | 0.90 |
| P(TfOMI/MI/t-BOCSt) | 1 | 0.33 | 3 | 89 | 0.90 |
| P(MsOMI/St) | 2 | 0.50 | 1 | 85 | 0.81 |
| P(MsOMI/t-BOCSt) | 2 | 0.50 | 1 | 94 | 0.72 |

[a] All of the copolymers have an alternating structure. Copolymerizations and terpolymerizations were carried out in 1:1 and 1:1:2 molar feed ratio, respectively, at 55°C in dioxane. [b] Mole % of the initiators AIBN to the combined monomers. [c] Ratio of the total monomer weight to the volume of dioxane. [d] Inherent viscosity measured at a concentration of 0.20 g/dl in dioxane at 25°C. [e] Molecular weights measured by GPC in a mixture of chloroform and THF (1:1 by vol), $M_W = 5.72 \times 10^5$. [f] $M_W = 5.80 \times 10^5$.

the polymers were controlled by adjusting the concentrations of the radical initiator and the monomers. The composition of the copolymers P(RsOMI/XSt) was confirmed to have the same molar ratio as the monomer feed ratio by NMR spectra and elemental analysis. The obtained copolymers are considered to have an alternating composition of the maleimide and styrene units since it is well-known that the radical copolymerization of an electron-poor monomer (maleimides) and an electron-rich monomer (styrenes) gives an alternating structure21-24).

## Thermal Properties of Polymers

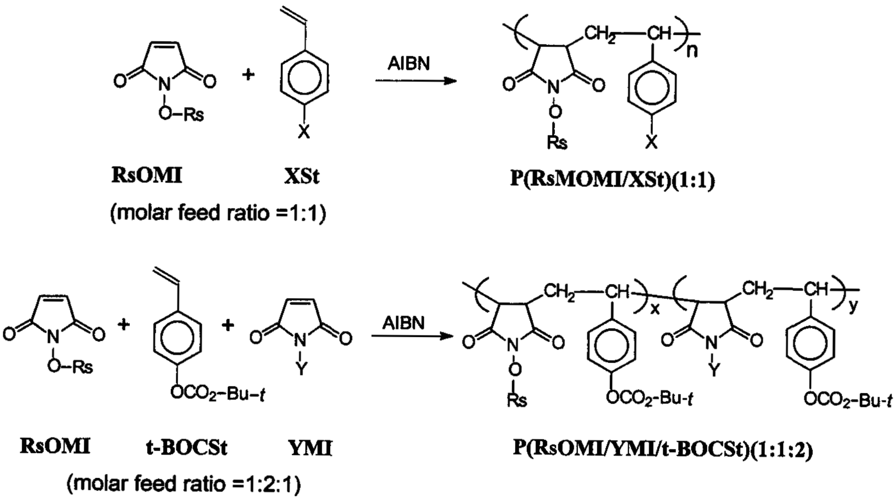
Thermal properties of P(RsOMI/XSt) were investigated by utilizing thermal analysis techniques, differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA). The DSC thermograms of the RsOMI polymers having t-BOCMI and/or t-BOCSt units revealed endothermic events in the range of about 150 °C to 180 °C due to the deprotection of t-BOC groups. In TGA thermograms, the amounts of the mass loss during the deprotection were found to be the same as the theoretically calculated amounts based on liberation of carbon dioxide and isobutylene (i.e. Fig. 2 and 3). The deprotection leads to the conversion of t-BOCMI and t-BOCSt units into MI and HOSt units, respectively, which was confirmed by FT-IR and $^1$H NMR spectral analyses. The deprotected polymers with MI and/or HOSt units showed high $T_g$s at above 200 °C.

The RsOMI polymers revealed extraordinary strong exothermic events beginning at about 220 °C for CSOMI, 225 °C for TfOMI, 260 °C for TsOMI, and 265 °C for MsOMI units in DSC analysis. The events are considered as cleavage of the corresponding sulfonyloxy groups from the polymers since the mass loss measured by the TGA thermograms nearly matches with the calculated amounts of the sulfonyloxy groups in each polymer. Immediately after the cleavage of sulfonyloxy groups the main-chains of the polymers eventually began to decompose.

## Photochemical Acid Generation16,18,20)

The photochemical generation of RsOH from P(RsOMI/XSt) was confirmed by isolation and identification of the acid. For example, the photogenerated camphorsulfonic acid (CSA) from P(CsOMI/HOSt) was isolated by solvent extraction from the UV-exposed polymer film and confirmed with $^1$H NMR spectral analysis. It was also verified by photolysis of *N*-(10-camphorsulfonyloxy)succinimide as a model compound, which generated CSA and succinimide20,25).

## Scheme 2



**RsOMI** **XSt** **P(RsMOMI/XSt)(1:1)**

(molar feed ratio =1:1)

**RsOMI** **t-BOCSt** **YMI** **P(RsOMI/YMI/t-BOCSt)(1:1:2)**

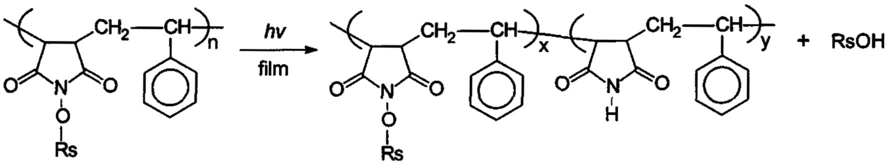
(molar feed ratio =1:2:1)

Rs = Ts, $CH_3SO_2$, $CF_3SO_2$, camphorsulfonyl

X = H (St), Me (MeSt), OH (HOSt), $OCO_2$-Bu-*t* (t-BOCSt), OAc (AcOSt)

Y = H (MI), OH (HOMI) , $C_6H_4OH$-*p* (HOPMI) ,$CO_2$-Bu-*t* (t-BOCMI)

## Scheme 3



**P(RsOMI/St)** **P(RsOMI/MI/St)**

Rs = Ts, $CH_3SO_2$, $CF_3SO_2$, camphorsulfonyl

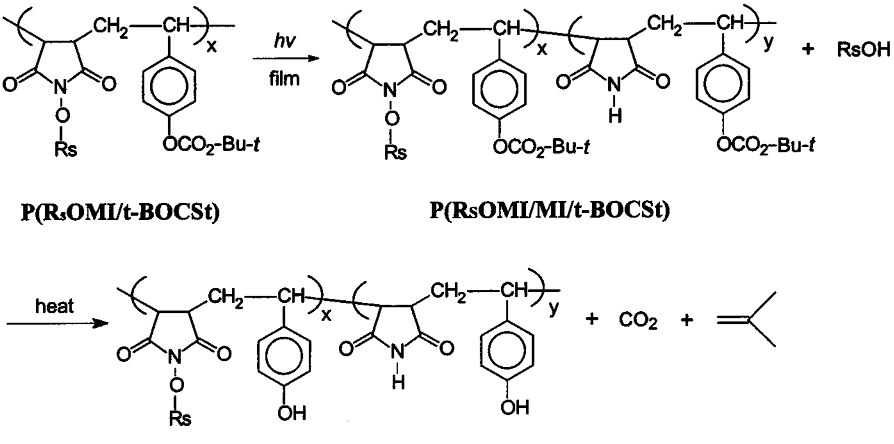
In photolysis of the P(RsOMI/XSt) films, not all of the RsOMI units yielded the corresponding sulfonic acid (RsOH) but only x mole fractions are converted into RsOH and MI units as described by the photolyzed polymer structure in Scheme 3. The amounts of RsOH generated by photolysis in film were determined by colorimetry utilizing a merocyanine dye or sodium salt of tetrabromophenol blue16,25,26). The percent conversions of CsOMI units in P(CsOMI/XSt) at various UV exposure doses are shown in Figure 1. The degree of photoconversion of P(CsOMI/St) was slightly higher than that of P(TsOMI/St) or P(MsOMI/St). When hydroquinone (HQ) was added as a hydrogen donor, the photochemical generation of CSA in P(CsOMI/St) films was significantly enhanced. The tendency of sensitivity enhancement by the use of HQ was also observed in other P(RsOMI/St). A photosensitized electron-transfer mechanism has been postulated in the photoinduced N-O bond cleavage of *N*-acyloxyphthalimides with subsequent sulfonic acid generation in the same manner from dicarboximido (succinimido) sulfonates12,13,27). Hydrogen donating additives like HQ in the photochemical reaction are known to enhance the capability of photochemical acid generation from the sulfonate PAGs13).

To provide the same enhancing effect as the HQ additive, HOSt units were incorporated into the CsOMI polymers by copolymerizing HOSt instead of St. The resulting phenolic polymer P(CsOMI/HOSt) revealed a remarkable increase in acid generation twice as much as that of P(CsOMI/St), and underwent photoconversion exceeding 70 mol % when irradiated with 1300 mJ/cm$^2$. P(CsOMI/HOSt) containing 5 wt % HQ exhibited the most effective acid generation among the CsOMI polymers with various combinations of comonomers and the HQ additive in this study.

## Deprotection of t-BOC Groups by Sulfonic Acids Generated *in situ*16-20)

Upon exposure to 250-nm UV with 75 mJ/cm$^2$, the RsOMI polymers having acid-labile t-BOC groups, P(RsOMI/t-BOCSt) showed a distinctive endothermic event owing to the deprotection of t-BOC groups at much lower temperatures in their DSC thermograms than the deprotection temperatures ($T_{dp}$) of the unexposed polymers, implying the acidolytic deprotection by the photogenerated RsOH as depicted in Scheme 4. When the RsOMI polymers P(RsOMI/t-BOCSt)(1:1) were photolyzed, x mole fractions of RsOMI units generated RsOH to give P[(RsOMI)$_{1-x}$/(MI)$_x$/t-BOCSt], leaving t-BOC groups intact. In subsequent heating the acidolytic deprotection afforded fully deprotected polymers P[(RsOMI)$_{1-x}$/(MI)$_x$/HOSt]. The acidolytic deprotection of side-chain t-BOC groups catalyzed by the photogenerated RsOH was confirmed by TGA in which the observed amounts of mass

Scheme 4



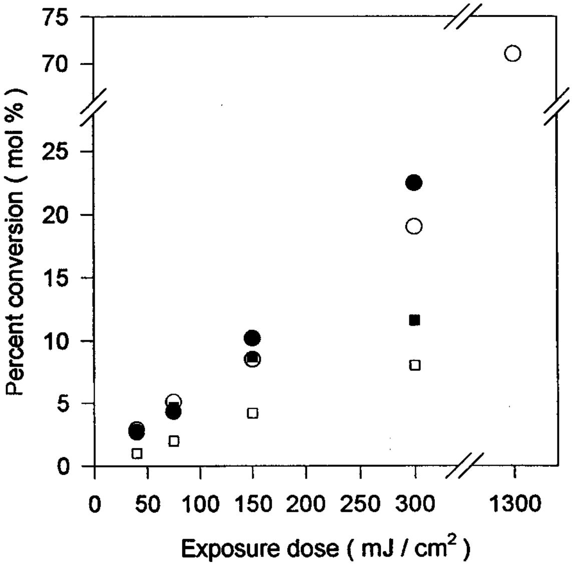


Fig. 1. Photochemical generation of camphorsulfonic acid from the CsOMI polymers by UV exposure in film: (○) P(CsOMI/HOSt); (●) P(CsOMI/HOSt) + 5% HQ; (□) P(CsOMI/St); (■) P(CsOMI/St) + 5% HQ.

loss caused by the deprotection at about the corresponding $T_{dp}$ are equal to the theoretically calculated amounts.

In TGA thermograms as shown in Fig. 2, P(TsOMI/t-BOCMI/t-BOCSt) and P(TsOMI/MI/t-BOCST) reveal the equal amounts of the mass loss with 33% and 25%, respectively, at the different $T_{dp}$s before and after the UV exposure in the films$^{17)}$. In Fig. 3 the TGA thermograms of P(CsOMI/t-BOCSt)(1:1) exhibit the same mass loss of 19% due to the complete deprotection of t-BOC groups by the photogenerated CSA after UV exposure$^{19)}$. Upon exposure to 250nm-UV with 75mJ/cm$^2$, P(CsOMI/t-BOCSt) showed distinctive endothermic events in its DSC thermograms by the deprotection at the different $T_{dp}$s as depicted in Fig. 4$^{20)}$. The acidolytic deprotection of P(CsOMI/t-BOCSt) was observed at 125°C quite lower than its original thermal deprotection temperature at 177°C without exposure. $T_{dp}$ decreased further to 91°C when 5% HQ was added to the polymer as clearly shown in Fig. 4. The thermal deprotection behavior of P(RsOMI/t-BOCSt)(1:1) is much related to the nature of photogenerated acids (RsOH) so that the deprotection temperatures significantly decrease with the increase in the order of the acid strength CSA<TsOH<MsOH<TfOH. The deprotection temperatures of the t-BOC groups were observed by DSC and are summarized in Table 3$^{17,20)}$. $T_{dp}$s decreased further when the polymers contained small amounts of HQ.

Table 3. Deprotection temperatures ($T_{dp}$) of t-BOC groups in the copolymers of RsOMI and t-BOCSt before and after exposure*

| Polymer | Unexposed (°C) | Exposed (°C) | Exposed with HQ# (°C) |
|---|---|---|---|
| P(CSOMI/t-BOCSt) (1:1) | 177 | 125 | 91 |
| P(TsOMI/t-BOCSt) (1:1) | 171 | 109 | 84 |
| P(TsOMI/MI/t-BOCSt) (1:1:2) | 171 | 116 | 93 |
| P(TsOMI/t-BOCMI/t-BOCSt) (1:1:2) | 171 | 120 | 94 |
| P(MsOMI/t-BOCSt)(1:1) | 166 | 110 | 60-90 |
| P(TfOMI/t-BOCSt) (1:1) | 150 | 87 | 79 |
| P(TfOMI/MI/t-BOCSt) (1:1:2) | 150 | 86 | 81 |

*Determined by DSC analysis before and after exposure to 250-nm UV with 75 mJ/cm$^2$ in film.
#Copolymers containing 10 % hydroquinone (HQ); 5 % HQ for terpolymers.

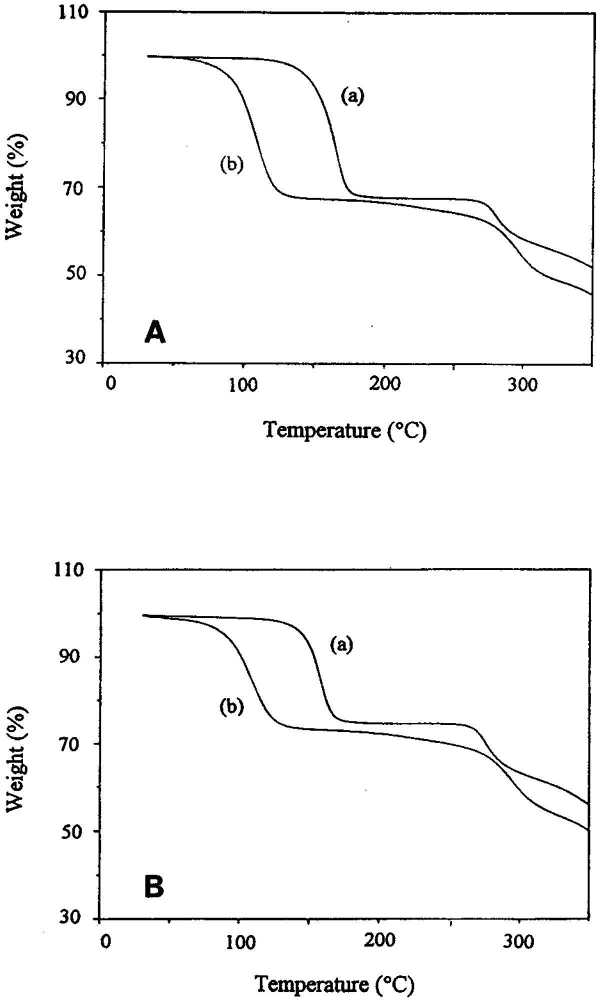


Fig. 2. TGA thermograms of (A) P(TsOMI/t-BOCMI/t-BOCSt) and (B) P(TsOMI/ MI/ t-BOCSt) (a) before and (b) after exposure to 250-nm UV with 75 mJ/cm$^2$ for TsOH generation and deprotection in film.

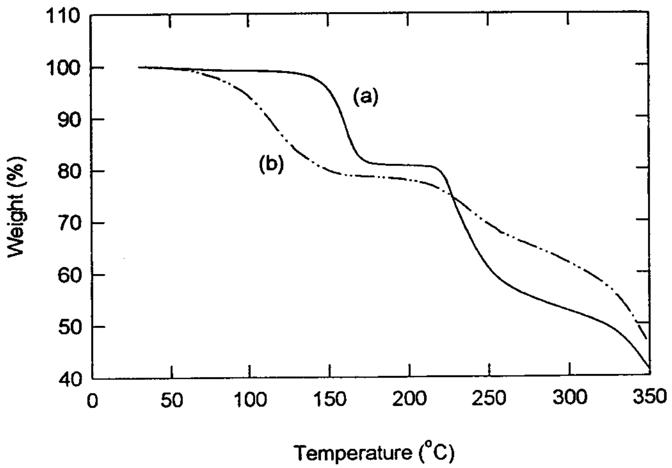


Fig. 3. TGA thermograms of P(CsOMI/t-BOCSt) (a) before and (b) after exposure to 250-nm UV with 100mJ/cm$^2$ for CSA generation and deprotection in film.

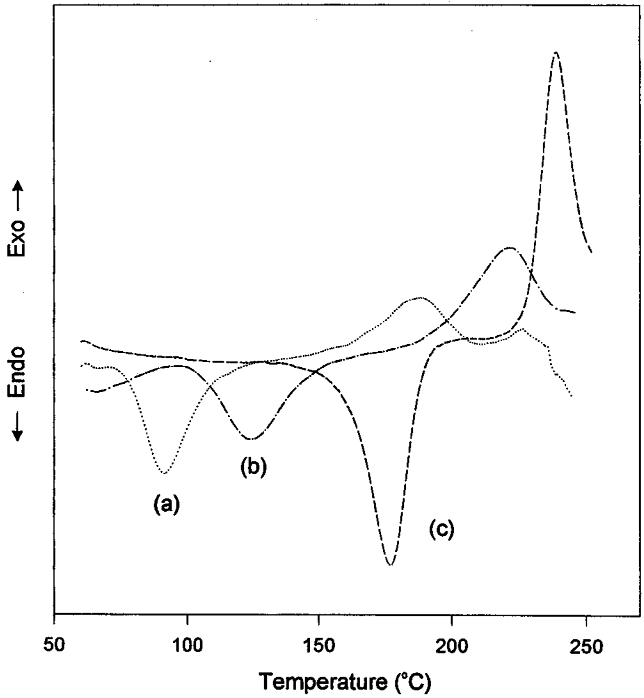


Fig. 4. DSC thermograms of P(CsOMI/t-BOCSt) after exposure to 250-nm UV with 75 mJ/cm$^2$: endothermic peaks appeared at (a) 91°C with 10% HQ, (b) 125 °C without HQ, (c) 177 °C without exposure.

### Application as Single-Component Photoresists[17,19,28-30)

The copolymers and terpolymers of RsOMI have a good film-forming property and relatively low optical densities (OD) less than 0.3/μm at 250 nm except TsOMI terpolymers which have ODs of about 0.4/μm. All of the copolymers P(RsOMI/t-BOCSt) are soluble in common organic solvents such as chloroform, cyclohexanone, THF, and anisole, but insoluble in aqueous alkaline solutions such as 1.0 N NaOH and 2.38 wt % tetramethylammonium hydroxide (TMAH). However, they showed a significant change in solubility after DUV exposure and post-exposure bake (PEB) due to the large polarity change: the deprotected polymers are soluble in aqueous base developers.

The RsOMI polymers were employed as a single-component, chemically amplified photoresist without adding any PAG. The photoresist films were imagewise exposed by a contact mode using a 250-nm exposure system with doses of 50 to 100 mJ/cm$^2$ and then baked in order to induce acidolytic deprotection in the exposed areas. Negative- and positive-tone patterns were obtained by developing with organic and aqueous alkaline developers, respectively. We have extended the concept utilizing the RsOMI polymers having t-BOC groups to new single-component photoresists consisting of *N*-vinylpyrrolidone[28,29), and t-BOC-norbornene and *N*-(10-camphorsulfonyloxy)norborneneimide[30).

### Photoinduced Functional Imaging

Selective transformation of chemical functionalities into the desired format is extremely worthwhile for making functional imaging onto thin polymer films[31-33). Photoinduced reactions in the aforementioned RsOMI polymers are simple and versatile means to achieve the desired selectivity between the exposed and unexposed areas, leading to functional transformation in the defined (exposed) area. Upon incorporation of both the RsOMI and the acid-labile t-BOC group into a single reactive polymer chain, the resulting RsOMI polymers are willing to be transformed into new functional forms by virtue of photoacid generation and subsequent acid-catalyzed deprotection only in the exposed area. We have selectively generated such functional groups as free imide NH from t-BOCMI units, $CO_2H$ from *t*-butyl ester moieties and phenolic OH from t-BOCSt units in the exposed polymer films.

Recently, we have prepared *N*-(10-camphosulfonyloxy)norborneneimide (CsNBI) and *N*-(tosyloxy)norborneneimide as functional norborneneimides ($R_f$NBI) and polymerized them to obtain new photoacid generating polymers[30). These polymers having acid-labile protecting groups are again applicable as single-component photoresists in lithography, and as new functional polymers in photoimaging and functional imaging.

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