Mechanical Property Investigation of Single Polymer Particles with the Variation of Molecular Structure of Crosslinking Monomer

Dong Ok Kim, Jeong Hee Jin

Advanced Materials Research Division, Hanwha Chemical, 6 Shinsung-Dong, Yusung-Gu, Daejon 305-804, South Korea

ABSTRACT: The effects of the swelling ratio (S/R), the existence of methyl side groups, the length and molecular structure of backbone chain of crosslinking monomer on the variation of mechanical properties of single polymer particle were investigated. For the study, monodisperse polymer particles were prepared via one-step seeded polymerization using PMMA as seed particles and 1,n-alkanediol di(meth)acrylate or (ethylene glycol)$_n$ di(meth)acrylate as crosslinking monomer. Recovery rate, K-values, breaking strength, and breaking displacement were measured as mechanical properties and they were performed by using microcompression test. The following observations were made: (1) only breaking strength was closely related to the swelling ratio; (2) the existence of methyl side groups increased the K-values but decreased the recovery rate of the polymer particles; and (3) K-values were the most sensitive to the variation of backbone chain length of crosslinking monomer, and they were decreased with the increase of the backbone chain length of crosslinking monomer. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 105: 783–789, 2007

Key words: polymer particle; swelling ratio; crosslinking monomer; molecular structure; mechanical property

INTRODUCTION

During the past decade, numerous investigators[1–13] reported on the new electronic packaging technology for the miniaturization and environmentally friendly manufacturing process of electronic devices. There are too many literatures to cite them all here, but the one of successful examples is the development of anisotropic conductive film (ACF) technology. Because it can replace conventional soldering and underfill encapsulation processes causing environmental problems and high cost, it is being widely used for packaging technology in flat panel display, such as tape carrier package,[1] chip on glass,[2] or chip on film,[3,4] and is extending its application field to flip chip packaging.[5–10]

ACFs are adhesive films with anisotropic conductivity induced by dispersing conductive particles into polymer matrices such as thermoplastics and thermosetting resins. Mostly metal-coated inorganic or organic particles are used as these conductive particles, and they usually have double layer structure that consists of Ni inner layer for electrical conductivity and Au outer layer for protecting inner layer from the oxidation and increasing the reliability of electrical performance.[11–13] However, micron-sized polymer particles with proper mechanical properties (elasticity, hardness, etc.) are preferred as core materials in these days because they can be deformed between bonding electrodes during thermal compression, and this increases the contact area between the bonding electrodes and conductive particles. It is helpful for lowering the connection resistance. Therefore, preparing micron-sized polymer particles having a wide range of mechanical properties is very important.

Traditionally, micron-sized polymer particles have been synthesized by suspension polymerization. However, in spite of the simplicity of suspension polymerization, the size distribution of polymer particles becomes very broad due to the intrinsic nature of suspension polymerization. Thus alternative methods for producing monodisperse micron-sized polymer particles have been developed, including (i) the successive seeding and polymerization (Vanderhoff et al.[14]), (ii) the activated swelling method (Ugelstadt and coworkers[15–17]), and (iii) the dynamic swelling method (Okubo and coworkers[18,19]). About swelling method, one- or two-steps are needed, i.e., seed polymer particles absorb monomer directly in one-step method, but they need to be activated by absorbing

Correspondence to: D. O. Kim (sky2000cokr@hotmail.com).

a water-insoluble compound such as 1-chlorododecane at first and then absorb monomer in two-steps method. In these methods, although the molecular structure of crosslinking monomer is one of the most important parameters for determining the mechanical properties of polymer particles, very little systematic investigation has been reported in the literature except for recently published Kim et al.'s study.[20] However, because their investigation was restricted to the mechanical properties of poly(MMA-HDDA) particle, it was not enough for systematic understanding of the relationship between molecular structure of crosslinking monomer and mechanical properties of polymer particles.

In this study, using five mechanical properties (i.e., recovery rate, K-values, breaking strength, and breaking displacement) that were introduced in previously published literatures,[20–23] we investigated the effects of the swelling ratio, the existence of methyl side groups, the length, and molecular structure of backbone chain of crosslinking monomer on the variation of mechanical properties of polymer particles prepared via one-step seeded polymerization.

EXPERIMENTAL

Materials

For the preparation of PMMA seed polymer via soap-free polymerization, MMA (99%, Junsei, Japan) was purified using an inhibitor removal column (Aldrich, USA) and stored at −5°C prior to use. KPS (potassium persulfate, Junsei) was used as initiator without further purification and 1-Octanethiol (Aldrich) was used as chain transfer agent.

BDDA (1,4-butanediol diacrylate), BDDMA (1,4-butanediol dimethacrylate), HDDA (1,6-hexanediol diacrylate), HDDMA (1,6-hexanediol dimethacrylate), diEGDMA [di(ethylene glycol) dimethacrylate], and triEGDMA [tri(ethylene glycol) dimethacrylate] were purchased from Aldrich and used as crosslinking monomer without further purification for the seeded polymerization. BPO (benzoylperoxide, Lancaster) was used as initiator. PVA (polyvinylalcohol, Aldrich, M_w: 85,000–146,000) was used as dispersant for seeded polymerization. SLS (sodiumlaurylsulfate, Aldrich) was used as emulsifying agent for emulsification of monomer mixture.

TABLE I
Standard Recipe for the Preparation of PMMA Seed Particles by Soap-Free Polymerization

Chemicals	Amount (g)
MMA	40
1-Octanethiol	0.5
KPS	0.3
Water	690

TABLE II
Physical Properties of Seed Polymer Particles

Diameter (μm)	0.84
C_v	5.6
M_W	19,400

$C_v(\%) = \sigma/D_n \times 100$, where σ, standard deviation of diameter; D_n, number average diameter.

Polymerizations

The polymerization procedure for the preparation of PMMA seed particles via soap-free polymerization is as follows. First, the chemicals summarized in Table I were put into a 1000-mL round flask equipped with a mechanical stirrer and reflux condenser under a well-controlled temperature. The chemicals were stirred gently with nitrogen purging for 1 h to remove dissolved oxygen. Then, the temperature of the reactor was increased to 70°C and a predetermined amount of initiator was added to the reactor, and the polymerization reaction was continued 8 h. The reactor was cooled down and the seed particles were separated using centrifugation, washed with deionized water three times, and dried in a vacuum oven. The characteristics of seed particles thus prepared are summarized in Table II.

For the seeded polymerization, the chemicals summarized in Table III were added to a 50 mL vial and emulsified using ultrasonic homogenizer. A predetermined amount of seed particles was added to above monomer emulsion and this vial was placed in a shaking incubator at room temperature for 24 h for monomer absorption. After the monomer absorption, a predetermined amount of PVA solution was added and polymerization was performed in a shaking bath under temperature control for 24 h. After the polymerization, the same separation and cleaning procedures employed for the seed polymerization were used to collect the poly(MMA-HDDA) particles.

Characterizations and mechanical property measurements

The analysis of polymer particle size and distribution was performed by using AccuSizer™ 780A (PSS.NICOMP, USA). The mechanical properties such as recovery rate, K-values, breaking strength,

TABLE III
Ingredients for the Synthesis of Polymer Particles by Seeded Polymerization

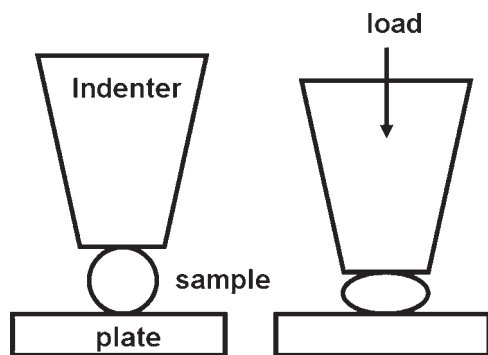| Chemicals	Amount (g)
Crosslinking monomer	1
Initiator	0.05
SLS (0.3%) solution	17
PVA (3%) solution	3

Figure 1 Schematic of the MCT.

and breaking displacement were obtained by calculating the average value of 10 individual particles' MCT (microcompression test, Fisher H100C) measurements. The measurement method for each mechanical property is as follows.

Recovery rate (R_r)

As shown in Figures 1 and 2(a), A polymer particle (diameter: D, radius: R) is placed on the smooth surface sample table and compressed with indenter at the speed of 1 mN/s to maximum load value (5 mN), and after that, the load is decreased at the same speed to standard load value (0.1 mN). The recovery rate represents the degree of particle recoverability after being deformed by compressing, and is defined as the ratio (L_2/L_1) represented by %, wherein L_1 is the displacement from standard load point to maximum load point during compression and L_2 is the displacement from the maximum load point to the standard load value during releasing.

$$R_r = [L_2/L_1] \times 100$$

K-values (K_{10} and K_{20})

Load value (F) and compression displacement (S) at the point of 10% (or 20%) compression deformation of the particle are measured while compressing single particle with indenter at the speed of 0.67 mN/s. K-values are defined as follows and represent the degree of particle hardness.

$$K = (3/2^{1/2})FS^{-3/2}R^{-1/2}$$

Breaking strength (S_0)

Breaking strength represents the compression rupture strength of the particle. It is calculated by measuring load value (Q) at the breaking point while compressing with indenter at the speed of 0.67 mN/s

as shown in Figure 2(b), and defined as follows.

$$S_0 = 2.8Q/\pi D^2$$

Breaking displacement (F_r)

Breaking displacement represents the compression rupture displacement of the particle. It is calculated by measuring compression displacement (L) at the breaking point while compressing as shown in Figure 2(b), and defined as the ratio (L/D) repre-
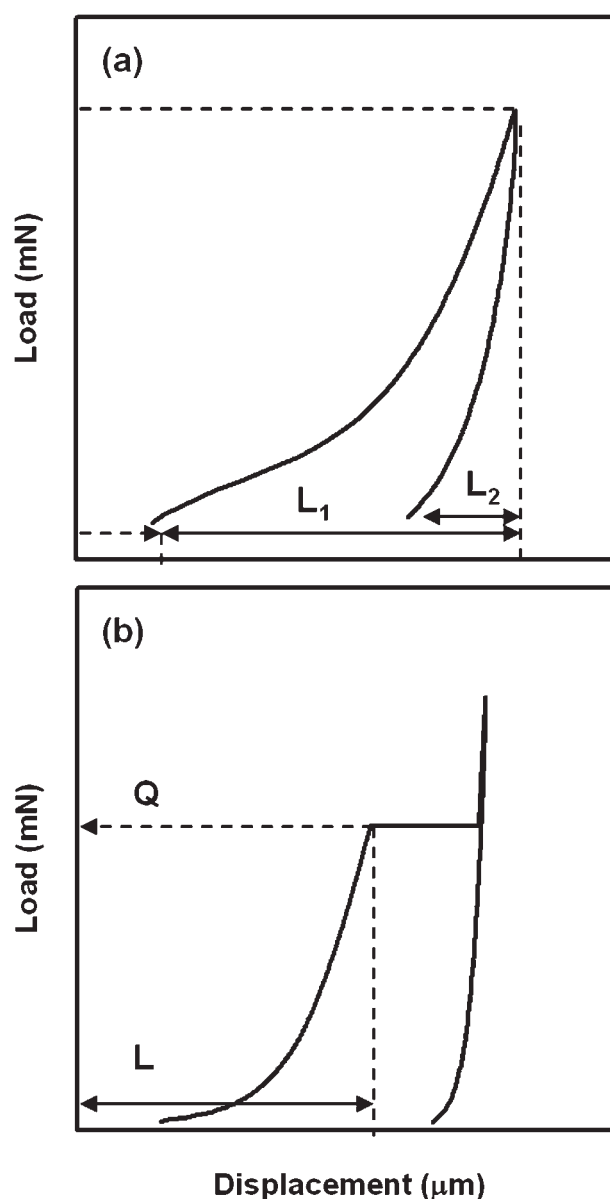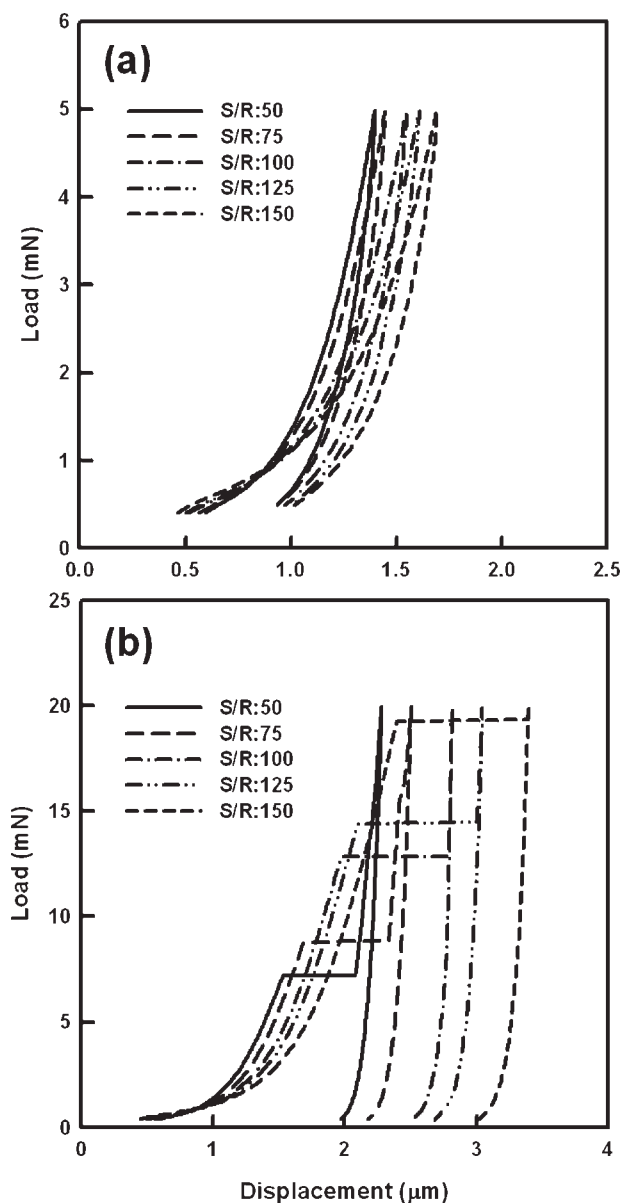

Figure 2 Plots of load versus compression displacement for (a) recovery rate measurement and for (b) K-values, breaking strength, and breaking displacement of monodisperse polymer particle.

Figure 3 Plots of load versus displacement for (a) recovery rate measurement and for (b) K-values, breaking strength, and breaking displacement of poly(MMA-BDDA) particles with different swelling ratios.

sented by %, wherein D is the diameter of particle.

$$F_r = (L/D) \times 100$$

RESULTS AND DISCUSSION

Effect of swelling ratio

Preparing the conductive particles having a wide range of mechanical properties can be achieved through the control of mechanical properties of core materials. In the case of seeded polymerization, mechanical properties of polymer particles are mainly

determined by several parameters, such as the compatibility of seed polymer with crosslinking monomer, molecular weight of seed polymer, molecular structure of crosslinking monomer, swelling ratio, etc. Therefore, the systematic investigation for the effect of molecular structure of the crosslinking monomer on the mechanical properties of polymer particles is very important. As a first step for the understanding of relationship between the molecular structure of crosslinking monomer and mechanical properties of polymer particles, we performed the investigation for the effect of swelling ratio on the mechanical properties of polymer particles.

Figure 3(a) shows the MCT results for the measurement of the recovery rate, and Figure 3(b) shows the MCT results for the measurement of the K-values, breaking strength, and breaking displacement of poly(MMA-BDDA) particles with the increase of swelling ratio. Mechanical properties calculated from Figure 3 were summarized in Table IV. It can be seen in Table IV that recovery rate, K-values (K_{10} and K_{20}), and breaking displacement were remained more or less constant with the increase of swelling ratio. In contrast, the breaking strength was increased (91%) as the swelling ratio was increased from 50 to 150, which suggested that only breaking strength is sensitive to the variation of swelling ratio.

Following the same procedure shown above, mechanical properties of polymer particles that were prepared by using BDDMA, HDDA, and HDDMA as crosslinking monomer were also measured and their results were summarized in Tables V–VII respectively. As observed from poly(MMA-BDDA) particles, only breaking strength was increased significantly but other mechanical properties were remained more or less unchanged with the increase of swelling ratio. Specially, at the swelling ratio of 150, poly(MMA-BDDMA) particle was not broken by the compression load up to 20 mN during the compression deformation, and it made measurements of breaking strength and breaking displacement impossible. On the basis of the observation made above, we conclude that only breaking strength is closely related to the swelling ratio.

TABLE IV
Variation of Mechanical Properties of Poly(MMA-BDDA) Particles with Swelling Ratio

S/R	D_n (µm)	R_r (%)	K_{10} (MPa)	K_{20} (MPa)	S_0 (MPa)	F_r (%)
50	3.0	60	1998	1509	674	50
75	3.4	62	1911	1560	676	50
100	3.9	61	1958	1568	778	52
125	4.0	60	1963	1583	811	53
150	4.2	59	2013	1585	952	56

TABLE V
Variation of Mechanical Properties of Poly(MMA-BDDMA) Particles with Swelling Ratio

S/R	D_n (μm)	R_r (%)	K_{10} (MPa)	K_{20} (MPa)	S_0 (MPa)	F_r (%)
50	3.1	40	2585	2088	646	53
75	3.5	44	2443	2005	666	52
100	3.9	47	2569	2091	777	56
125	4.1	47	2614	2077	887	59
150	4.3	47	2730	2148	—	—

TABLE VII
Variation of Mechanical Properties of Poly(MMA-HDDMA) Particles with Swelling Ratio

S/R	D_n (μm)	R_r (%)	K_{10} (MPa)	K_{20} (MPa)	S_0 (MPa)	F_r (%)
50	3.1	40	2391	1841	724	55
75	3.4	42	2324	1768	791	57
100	3.7	45	2304	1791	850	58
125	3.8	45	2263	1755	833	58
150	3.9	46	2463	1900	948	62

Effect of side group

Although BDDMA has the same backbone structure as BDDA (i.e., butanediol group), it has two methyl side groups. Thus the comparison of mechanical properties of poly(MMA-BDDMA) particles with those of poly(MMA-BDDA) particles gives the information about the effect of side groups of crosslinking monomer on the variation of mechanical properties of polymer particles. From the comparison of Table V with Table IV, the recovery rate of poly (MMA-BDDMA) particle was lower (e.g., 24% lower at $S/R = 100$), whereas K-values were higher (e.g., 32% higher at $S/R = 100$) than those of poly(MMA-BDDA) particles throughout the entire range of swelling ratio. However, breaking strength and breaking displacement were about the same levels as those of poly(MMA-BDDA) particles.

As supplementary examples, the mechanical properties of poly(MMA-HDDMA) particles shown in Table VII were also compared with those of poly (MMA-HDDA) particles shown in Table VI. As observed from poly(MMA-BDDMA) particles, the recovery rate of poly(MMA-HDDMA) particle was lower (e.g., 28% lower at $S/R = 100$), while K-values were higher (e.g., 45% higher at $S/R = 100$) than those of poly(MMA-HDDA) particles throughout the entire range of swelling ratio. Breaking strength and breaking displacement were about the same levels as those of poly(MMA-HDDA) particles. These observations seem to suggest that the methyl side groups do not influence the mechanical properties measured at the breaking point (e.g., breaking strength and breaking displacement) significantly but have a great

TABLE VI
Variation of Mechanical Properties of Poly(MMA-HDDA) Particles with Swelling Ratio

S/R	D_n (μm)	R_r (%)	K_{10} (MPa)	K_{20} (MPa)	S_0 (MPa)	F_r (%)
50	3.2	63	1430	1098	670	54
75	3.7	63	1518	1271	743	51
100	4.0	64	1554	1257	804	54
125	4.2	61	1531	1228	881	53
150	4.3	56	1436	1206	1002	60

influence on the mechanical properties measured during the compression deformation (e.g., recovery rate and K-values).

To the best of our knowledge, we are not aware of any previous study that reported on the effect of side groups of crosslinking monomer on the mechanical properties of single polymer particle. Furthermore, we have no our own theory predicting the mechanical property variation of single polymer particle with the side groups. On the basis of the above observations, we just speculate that the existence of methyl side groups provides the steric hindrance against the polymer chain movement during compression deformation and gives rise to the increase of K-values, while it also provides steric hindrance against the polymer chain movement during compression releasing and induces the decrease of recovery rate. It is well documented[24] that bulky side groups play a significant role in determining the rheological behaviors (e.g., increasing the overshoot values of transient shear flow and the melt viscosity of steady shear flow or decreasing the stress relaxation rate) by providing the steric hindrance against the polymer chain movement, and the effect of bulky side groups on the rheological properties increases as the van der Waals volume of bulky side groups increases. Thus, we can conclude that introducing the bulky side group to the crosslinking monomer is very effective method to increase the K-values but decrease the recovery rate at the same time.

Effect of backbone chain length

Although BDDA has the same backbone structure as HDDA (i.e., alkanediol group), it has a shorter backbone chain (butanediol group) than HDDA (hexanediol group). Thus the comparison of mechanical properties of poly(MMA-BDDA) particles with those of poly(MMA-HDDA) particles provides the information about the effect of the backbone chain length of crosslinking monomer on the mechanical properties of polymer particles. From the comparison of Table IV with Table VI, the recovery rate, breaking strength, and breaking displacement of poly(MMA-BDDA) particles were about the same
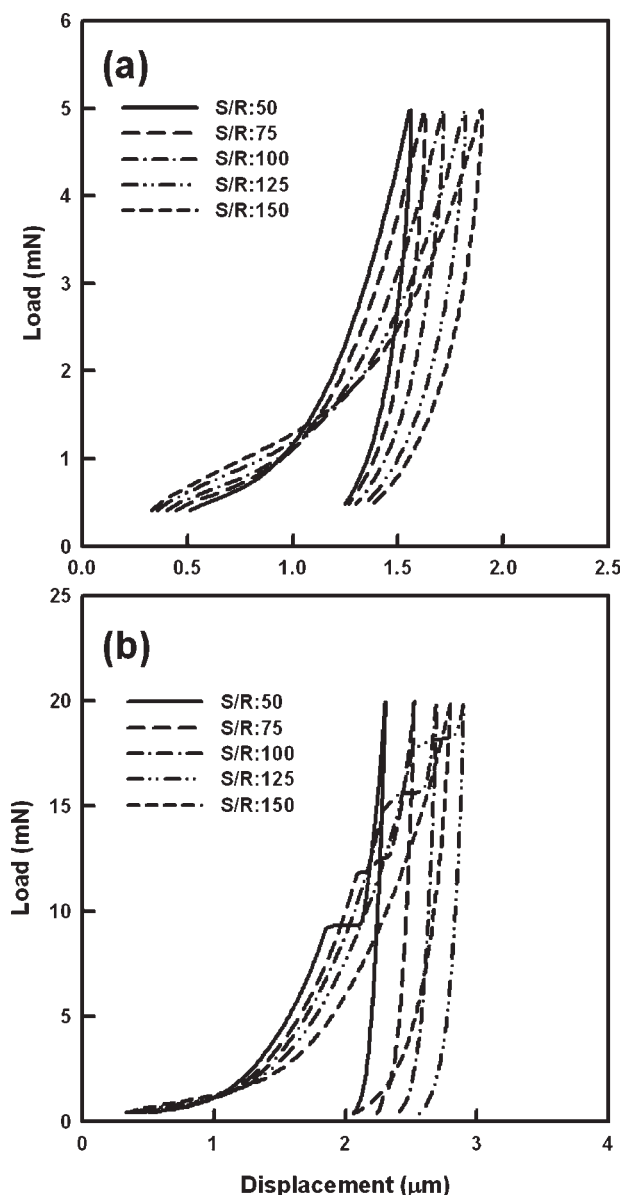
Figure 4 Plots of load versus displacement for (a) recovery rate measurement and for (b) K-values, breaking strength, and breaking displacement of poly(MMA-diEGDMA) particles with different swelling ratios.

levels as those of poly(MMA-HDDA) particles, while both K_{10} and K_{20} of poly(MMA-BDDA) particles were higher throughout the entire range of swelling ratio. Similar results were also observed from the comparison of poly(MMA-BDDMA) particles with poly (MMA-HDDMA) particles. For example, K_{10} and K_{20} of poly(MMA-BDDMA) particles at $S/R = 100$ were 20 and 14% higher, respectively. On the other hand, breaking strength of poly(MMA-BDDMA) particles remained more or less lower level than that of poly (MMA-HDDMA) particles until the swelling ratio reached 100, but it became higher after that. It is needless to say that decreasing the alkanediol back-

bone chain length of crosslinking monomer increased the polymer chain stiffness, and it induced the increase of hardness (K-values) of polymer particles.

According to the literatures,[25–29] when the polymer chains have methylene groups ($-CH_n-$) of flexible spacer, the stiffness of polymer chains having even numbers of methylene groups of flexible spacer are higher than those of polymer chains having odd numbers of flexible spacer, and it increases as the flexible spacer length decreases. Thus, the observations made above are in good agreement with those reported previously. However, more systematic experimental investigations on the relationship between backbone chain length of crosslinking monomer and mechanical properties of polymer particles are needed, to establish a firm scientific basis, on which meaningful theory can be developed.

Effect of backbone molecular structure

In addition to crosslinking monomers (BDDA, BDDMA, HDDA, or HDDMA) having alkanediol backbone chains, we also investigated the mechanical properties of polymer particles prepared by using diEGDMA or triEGDMA having (ethylene glycol)$_n$ ($n = 2$ or 3) backbone chains.

Figure 4(a) describes the MCT results for the measurement of the recovery rate, and Figure 4(b) describes the MCT results for the measurement of the K-values, breaking strength, and breaking displacement of poly(MMA-diEGDMA) particles with the increase of swelling ratio. Mechanical properties calculated from Figure 4 were summarized in Table VIII. As can be seen in Table VIII, all the mechanical properties were remained more or less unchanged with the increase of swelling ratio. However, as the swelling ratio was increased from 125 to 150, particles were not broken by the compression load up to 20 mN during the compression deformation as observed from poly(MMA-BDDMA) particles. On the other hand, recovery rate showed no significant change, while K-values were increased slightly. Mechanical properties of poly(MMA-triEGDMA) particles summarized in Table IX showed very similar results to those of poly(MMA-diEGDMA) particles.

TABLE VIII
Variation of Mechanical Properties of Poly(MMA-diEGDMA) Particles with Swelling Ratio

S/R	D_n (μm)	R_r (%)	K_{10} (MPa)	K_{20} (MPa)	S_0 (MPa)	F_r (%)
50	3.0	31	2681	1888	900	62
75	3.4	32	2651	1871	881	61
100	3.8	34	2469	1763	907	60
125	4.1	33	2640	1782	935	61
150	4.3	32	2918	1963	—	—

**TABLE IX
Variation of Mechanical Properties of Poly(MMA-triEGDMA) Particles with Swelling Ratio**

S/R	D_n (μm)	R_r (%)	K_{10} (MPa)	K_{20} (MPa)	S_0 (MPa)	F_r (%)
50	3.1	34	2185	1421	856	62
75	3.4	35	2066	1428	825	59
100	3.9	35	2012	1406	874	60
125	4.1	34	2182	1493	1002	64
150	4.4	33	2445	1603	—	—

From the comparison of mechanical properties of poly(MMA-diEGDMA) particles with those of poly(MMA-triEGDMA) particles, K-values of poly(MMA-diEGDMA) were more or less higher throughout the entire range of swelling ratio, whereas recovery rate, breaking strength, and breaking displacement of poly(MMA-diEGDMA) particles were about the same levels as poly(MMA-triEGDMA) particles. These observations are in good agreement with the observation made from poly(MMA-BDDMA) and poly(MMA-HDDMA) particles, driving us to conclude that the chain length of (ethylene glycol)$_n$ backbone as well as alkanediol backbone plays an important role in determining the K-values of polymer particles.

CONCLUSIONS

In this study we have observed the variations of mechanical properties of monodisperse polymer particles prepared via one-step seeded polymerization with the swelling ratio, the existence of methyl side groups, the length, and molecular structure of backbone chain of crosslinking monomer.

In the case of polymer particles prepared by using BDDA, BDDMA, HDDA, and HDDMA as crosslinking monomers, only breaking strength was increased significantly, while other mechanical properties were remained more or less constant with the increase of swelling ratio. Moreover polymer particles prepared by using diEGDMA and triEGDMA as crosslinking monomer also showed the similar behaviors to above polymer particles. Thus, we concluded that breaking strength is the most sensitive mechanical property to the variation of swelling ratio. The comparison of poly(MMA-BDDA) particles with poly(MMA-BDDMA) particles [or poly(MMA-HDDA) particles with poly(MMA-HDDMA) particles] showed that the existence of methyl side groups generates the steric hindrance against the polymer chains movement; therefore, it gives rise to the increase of K-values during compression but decrease of recovery rates during releasing.

Moreover, on the basis of the comparison of poly(MMA-BDDMA) particles with poly(MMA-HDDMA) particles [and poly(MMA-diEGDMA) with poly(MMA-triEGDMA)], we observed that K-values are the most sensitive mechanical properties to the variation of backbone chain length of crosslinking monomer independent of swelling ratio, and they were decreased with the increase of the backbone chain length of crosslinking monomer.

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