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### SYNTHESIS AND POLYMERIZATION OF ISOPROPENYL-1,3,5-TRIAZINES CARRYING A LONG ALKYL CHAIN WITH OXYETHYLENE SPACER AND THE SIDE CHAINS CRYSTALLIZATION OF OBTAINED COMB-LIKE POLYMERS

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## SYNTHESIS AND POLYMERIZATION OF ISOPROPENYL-1,3,5-TRIAZINES CARRYING A LONG ALKYL CHAIN WITH OXYETHYLENE SPACER AND THE SIDE CHAINS CRYSTALLIZATION OF OBTAINED COMB-LIKE POLYMERS

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### ABSTRACT

Isopropenyltriazines carrying a long alkyl group with oxyethylene spacer were synthesized from 2-amino-4-anilino-6-isopropenyl-1,3,5-triazine (AAIT) with chlorinated oligoethyleneglycol mono-octadecylethers in the presence of potassium hydroxide. The numbers of oxyethylene unit were 1, 2, and 3. Polymers could be readily obtained by polymerization of these monomers in benzene using 2,2'-azobisisobutyronitrile as an initiator. Glass transition temperature ( $T_g$ ) of obtained polymers was around 100°C. Copolymerization of these monomers with styrene was carried out and the monomer reactivity ratios were determined. The monomer reactivity was independent of the length of oxyethylene spacer. The crystallization of the alkyl side chains in the comb-like polymers was investigated by differential scanning calorimetry (DSC). The long alkyl side chains in the resulting comb-like polymers could be crystallized below the  $T_g$  of polymers. The crystallinity of the side chains evaluated by DSC increased by the introduction of flexible oxyethylene spacer into each monomer unit and with increasing the length of oxyethylene spacer.

*Key Words:* Isopropenyltriazine; Long alkyl side chain; Radical polymerization; Comb-like polymer; Copolymerization parameters; Oxyethylene spacer; Side chains crystallization.

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## INTRODUCTION

Polymers containing long alkyl groups as side chains, so-called comb-like polymers, exhibit aggregation of side chains similarly to graft copolymers and side-chain liquid crystalline polymers. The long alkyl side chains of the comb-like polymers can be crystallized independently of the stereoregularity of the main chain [1-7].

In previous papers, we reported the synthesis and polymerization of isopropenyl-1,3,5-triazines fitted with a long alkyl group, and found that the long alkyl side chains could be crystallized below the glass transition temperature ( $T_g$ ) of the main chain [8-11]. The crystallization and melting of the side chains in the glassy state of the main chain can be expected to serve as an application for functionality, such as a dramatic change in mechanical properties and control of gas permeability. The melting temperature and degree of crystallinity of the side chain of obtained polymers, however, were lower than those of conventional comb-like polymers such as poly(*n*-alkyl methacrylates), poly(*p-n*-alkylstyrenes), and poly(*N-n*-alkylacrylamides) due to the rigidity of the main chain to prevent the arrangement of side chains. The introduction of a flexible spacer between a main chain and alkyl group should be effective in order to increase the crystallinity of side chains in a similar manner as side-chain liquid crystalline polymers.

2-amino-4-anilino-6-isopropenyl-1,3,5-triazine (**AAIT**) can carry a substituent in nitrogen atom of anilino group selectively. In this work, monomers having a long alkyl group with an oligo(oxyethylene) unit as a flexible spacer were synthesized from **AAIT**. Comb-like polymers with a spacer could be prepared by free radical polymerization. The influence of the number of oligo(oxyethylene) unit on the side chains crystallization of the resulting comb-like polymers are investigated.

## EXPERIMENTAL

### Materials

2-amino-4-anilino-6-isopropenyl-1,3,5-triazine (**AAIT**) was prepared by the reaction of 1-phenylbiguanide with methacryloyl chloride [12]. Oligoethylene-glycol mono-octadecylethers were prepared from oligoethyleneglycols with 1-bromo-octadecane in the presence of potassium hydroxide at 150°C for 36 hours. Solvents and styrene were purified by distillation after appropriate drying. 2,2'-azobisisobutyronitrile (AIBN) was recrystallized from methanol before use. Other reagents were obtained commercially and used without further purification.

### Synthesis of 2-Amino-4-[*N*-(3-oxahenicosyl)anilino]-6-isopropenyl-1,3,5-triazine (**1**)

To a mixture of **AAIT** (38.6 g, 0.17 mol), dimethylformamide (DMF) (400 mL), and 1-chloro-3-oxahenicosane (66.8 g, 0.20 mol) prepared by chlorination of

ethyleneglycol mono-octadecylether with thionyl chloride, potassium hydroxide (14.2 g, 0.25 mol) was added and further stirred for 72 hours at 60°C. The reaction mixture was poured into water, and the precipitated solid was filtered. The crude product was recrystallized from methanol and then purified by column chromatography on silica gel (eluent: hexane/acetone = 9/1). The yield was 11.3 g (13%), mp 43°C.

$^1\text{H}$  NMR (in  $\text{CDCl}_3$ ):  $\delta$  = 0.87 (t, 3H), 1.25 (m, 30H), 1.51 (t, 2H), 2.05 (s, 3H), 3.40 (t, 2H), 3.68 (t, 2H), 4.15 (t, 2H), 5.01 (s, 2H), 5.45 (s, 1H), 6.30 (s, 1H), 7.24-7.47 ppm (m, 5H).

Analysis. Calcd for  $\text{C}_{32}\text{H}_{53}\text{N}_5\text{O}$ : C, 73.38; H, 10.20; N, 12.17%. Found: C, 73.54; H, 10.22; N, 12.17%.

### Synthesis of 2-Amino-4-[N-(3,6-dioxatetracosyl)anilino]-6-isopropenyl-1,3,5-triazine (2)

To a mixture of **AAIT** (34.1 g, 0.15 mol), dimethylformamide (DMF) (400 mL), and 1-chloro-3,6-dioxatetracosane (67.9 g, 0.18 mol) prepared by chlorination of diethyleneglycol mono-octadecylether with thionyl chloride, potassium hydroxide (12.3 g, 0.22 mol) was added and further stirred for 72 hours at 60°C. The reaction mixture was poured into water, and the precipitated solid was filtered. The crude product was recrystallized from ethanol and then purified by column chromatography on silica gel (eluent: hexane/acetone = 4/1). The yield was 14.7 g (17%), mp 43°C.

$^1\text{H}$  NMR (in  $\text{CDCl}_3$ ):  $\delta$  = 0.86 (t, 3H), 1.26 (m, 30H), 1.55 (t, 2H), 2.03 (s, 3H), 3.41 (t, 2H), 3.54 (m, 4H), 3.74 (t, 2H), 4.14 (t, 2H), 5.36 (s, 2H), 5.42 (s, 1H), 6.28 (s, 1H), 7.21-7.45 ppm (m, 5H).

Analysis. Calcd. for  $\text{C}_{34}\text{H}_{57}\text{N}_5\text{O}_2$ : C, 71.92; H, 10.12; N, 12.33%. Found: C, 72.04; H, 10.20; N, 12.34%.

### Synthesis of 2-Amino-4-[N-(3,6,9-trioxaheptacosyl)anilino]-6-isopropenyl-1,3,5-triazine (3)

To a mixture of **AAIT** (34.1 g, 0.15 mol), dimethylformamide (DMF) (400 mL), and 1-chloro-3,6,9-trioxaheptacosane (75.8 g, 0.18 mol) prepared by chlorination of triethyleneglycol mono-octadecylether with thionyl chloride, potassium hydroxide (12.8 g, 0.23 mol) was added and further stirred for 72 hours at 60°C. The reaction mixture was poured into water, and the precipitated solid was filtered. The crude product was recrystallized from ethanol and then purified by column chromatography on silica gel (eluent: hexane/acetone = 7/3). The yield was 17.1 g (19%), mp 42°C.

$^1\text{H}$  NMR (in  $\text{CDCl}_3$ ):  $\delta$  = 0.88 (t, 3H), 1.26 (m, 30H), 1.57 (t, 2H), 2.03 (s, 3H), 3.43 (t, 2H), 3.60 (m, 8H), 3.73 (t, 2H), 4.15 (t, 2H), 5.22 (s, 2H), 5.43 (s, 1H), 6.29 (s, 1H), 7.22-7.46 ppm (m, 5H).

Analysis. Calcd. for  $C_{36}H_{61}N_5O_3$ : C, 70.60; H, 10.05; N, 11.44%. Found: C, 70.41; H, 10.10; N, 11.24%.

### Polymerization

Polymerization initiated by AIBN was carried out in benzene in a degassed sealed tube at 60°C. After the polymerization, its content was poured into a large amount of ethanol to precipitate the polymer. The resulting polymer was purified by reprecipitation from benzene with ethanol. The copolymer composition was evaluated from the elementary analysis of carbon and nitrogen.

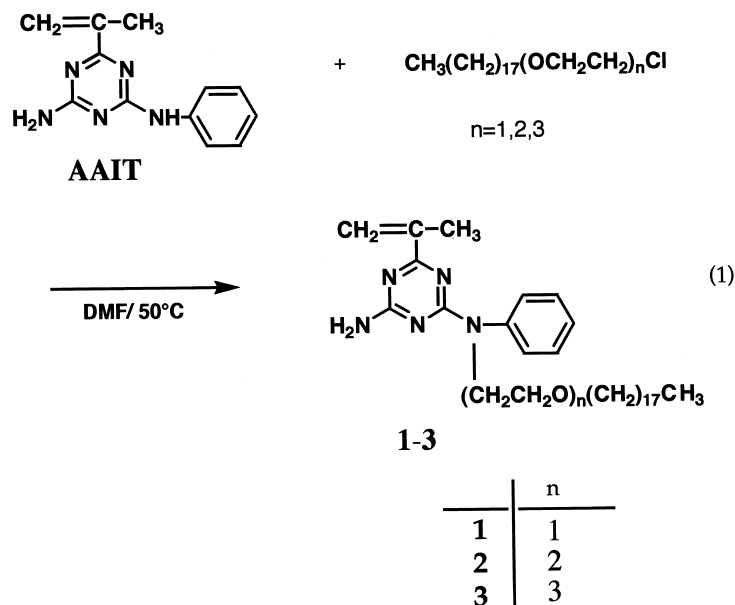
### Measurements

$^1H$  NMR spectra were recorded by a JEOL JNM-GX400 (400 MHz) spectrometer with  $CDCl_3$  as a solvent using tetramethylsilane as the internal standard. IR spectra were measured by a Nicolet Impact-400D. Gel permeation chromatography (GPC) was performed on a Tosoh DP-8020 with G2000, G3000, and GMH TSK gel-columns and differential refractometric detector using tetrahydrofuran (THF) as eluent at 40°C. Molecular weights were determined using molecularly uniform polystyrene standards. Differential scanning calorimetry (DSC) was performed on a Rigaku Denki DSC-8230 instrument at heating and cooling rates of 10°C/min. Indium and benzoic acid were used as standards for calibration of temperature and heat energy. Elementary analysis was carried out with Yanako CHN corder MT-3.

## RESULTS AND DISCUSSION

We have already reported the synthesis of 2-amino-4-(*N*-alkylanilino)-6-isopropenyl-1,3,5-triazines by the reaction of 2-amino-4-anilino-6-isopropenyl-1,3,5-triazine (**AAIT**) with alkyl halides in the presence of a base such as sodium hydride and potassium hydroxide in dipolar aprotic solvent [8, 13-15]. The reaction proceeded selectively through the anilide anion of **AAIT**, which could be followed by the  $^1H$  NMR measurements. The reaction selectivity would be due to the difference in stability between anilide anion of anilino group and amide anion of amino group.

By analogy with the above reaction, isopropenyltriazine monomers carrying a long alkyl chain with oligo(oxyethylene) spacer between triazine ring and alkyl group were synthesized from **AAIT** with end chlorinated oligoethyleneglycol mono-octadecylethers in the presence of potassium hydroxide as described in Equation 1. The numbers of oxyethylene units were 1 (**1**), 2 (**2**), and 3 (**3**).



Homopolymerization of these monomers **1-3** by AIBN in benzene at 60°C was carried out, and the results are summarized in Table 1. Polymers (**P-1-3**) could be obtained with high molecular weight, 285,000-398,000. The glass transition temperatures ( $T_g$ ) of the resulting polymers were observed at around 100°C determined by DSC measurement. The  $T_g$  decreases with increasing the number of oxyethylene unit.  $T_g$  of poly(2-amino-4-(*N*-octadecylanilino)-6-isopropenyl-1,3,5-triazine) (poly**N18**) was 132°C as reported previously [8]. This result suggested the increase in flexibility of polymer by increase in the length of oxyethylene spacer.

In order to estimate the reactivity of these monomers in radical copolymerization, copolymerization of **1-3** ( $M_2$ ) with styrene ( $M_1$ ) in benzene at various compositions of monomer feed was carried out at 60°C using AIBN as the initiator. The copolymerization was also carried out for 2-amino-4-(*N*-octadecylanilino)-6-isopropenyl-1,3,5-triazine (**N18**) as the monomer without oxyethylene

**Table 1.** Homopolymerization of Isopropenyl-1,3,5-triazines Initiated by AIBN at 60°C<sup>a</sup>

Monomer	Yield, %	$\eta_{sp}/c^b$ , dL/g	$M_n$	$M_w/M_n$	$T_g$ , °C	Sample
<b>1</b>	70	2.37	285,000	4.16	112	<b>P-1</b>
<b>2</b>	48	2.14	348,000	3.14	103	<b>P-2</b>
<b>3</b>	69	2.09	398,000	3.05	96	<b>P-3</b>

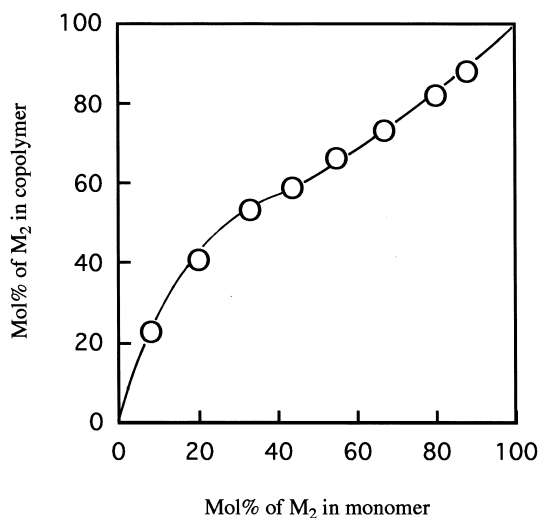
<sup>a</sup>Conditions: [monomer] = 0.4 mol/L, [AIBN] = 1.35 mmol/L, solvent = benzene, time = 90 minutes.

<sup>b</sup> $c$  = 0.2 g/dL in tetrahydrofuran

spacer in the same condition as above. The polymer yield was kept below 10% to evaluate the copolymerization parameters. The composition of the copolymers was determined by elementary analysis of carbon-nitrogen ratio. Figure 1 shows the monomer-copolymer composition curve of monomer **1** ( $M_2$ ). The composition curves of monomers **2**, **3**, and **N18** were almost same as that of **1**.

Monomer reactivity ratios ( $r_1, r_2$ ) in the above copolymerizations were estimated by the method of Kelen and Tüdös [16], and the resonance stabilization factor  $Q$  and electrical factor  $e$  values by Alfrey-Price were also calculated. They are summarized in Table 2 together with the previous values for 2-amino-4-(*N*-docosylanilino)-6-isopropenyl-1,3,5-triazine (**N22**) [17]. As shown in Table 2, no significant difference in the monomer reactivity ratios and Alfrey-Price  $Q$  and  $e$  values for the present monomers. The introduction of oxyethylene spacer did not affect the monomer reactivity.

The crystallinity of long alkyl side chains in the resulting comb-like polymers was investigated by means of differential scanning calorimetry (DSC). The measurements were carried out repeatedly from  $-120^\circ\text{C}$  to  $+150^\circ\text{C}$  at heating and cooling rates of  $10^\circ\text{C}/\text{min}$ . The second heating run was used for the evaluation of the crystallinity of the side chains, because the polymers have the same thermal history in DSC measurement. DSC curves of the second heating runs for **P-1**, **P-2**, and **P-3** are shown in Figure 2. Endothermic peaks ascribed to the melting of crystallized alkyl side chains were observed in the range of  $-50^\circ\text{C}$  to  $+20^\circ\text{C}$ . Melting temperature ( $T_m$ ) and heat of fusion ( $\Delta H_m$ ) of the side chain crystals were estimated from peak position and peak area, respectively. These values are shown in Table 3, together with the previous result for poly**N18** as the comb-like polymer



**Figure 1.** Monomer-copolymer composition curve of **1** ( $M_2$ ) with styrene ( $M_1$ ).

**Table 2.** Copolymerization Parameters of the Copolymerization of Isopropenyl-1,3,5-triazines ( $M_2$ ) with Styrene ( $M_1$ ) in Benzene at at 60°C

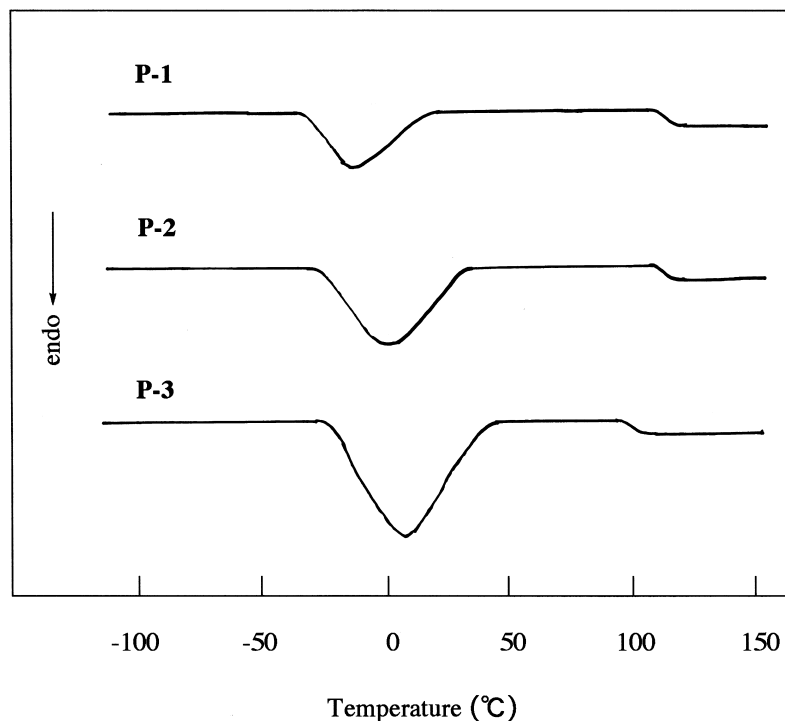
$M_2$	$r_1$	$r_2$	$Q_2$	$e_2$
<b>1</b>	0.16	0.72	1.93	0.67
<b>2</b>	0.16	0.79	1.98	0.64
<b>3</b>	0.16	0.75	1.86	0.64
<b>N18<sup>a</sup></b>	0.17	0.63	1.78	0.69
<b>N22<sup>b</sup></b>	0.20	0.55	1.53	0.69

<sup>a</sup>2-Amino-4-(*N*-octadecylanilino)-6-isopropenyl-1,3,5-triazine.

<sup>b</sup>2-Amino-4-(*N*-docosylanilino)-6-isopropenyl-1,3,5-triazine [13].

without spacer [8]. As seen in Figure 2, DSC curves of polymers showed  $T_g$  at around 100°C. The long alkyl side chains of the resulting comb-like polymers could be crystallized below the  $T_g$  of the main chain.

It is well known that the alkyl side chains of conventional comb-like polymers crystallize in hexagonal form [2-7]. Actually, the IR spectra of the present comb-like polymers measured at 10°C showed a single absorption peak at 720  $\text{cm}^{-1}$ , corresponding to the C-H vibration of the methylene chain, which indicate that the



**Figure 2.** DSC curves of P-1, P-2, and P-3 at a heating rate of 10°C/minutes.



**Table 3.** Thermodynamics Parameters of Polyisopropenyl-1,3,5-triazines Determined by DSC

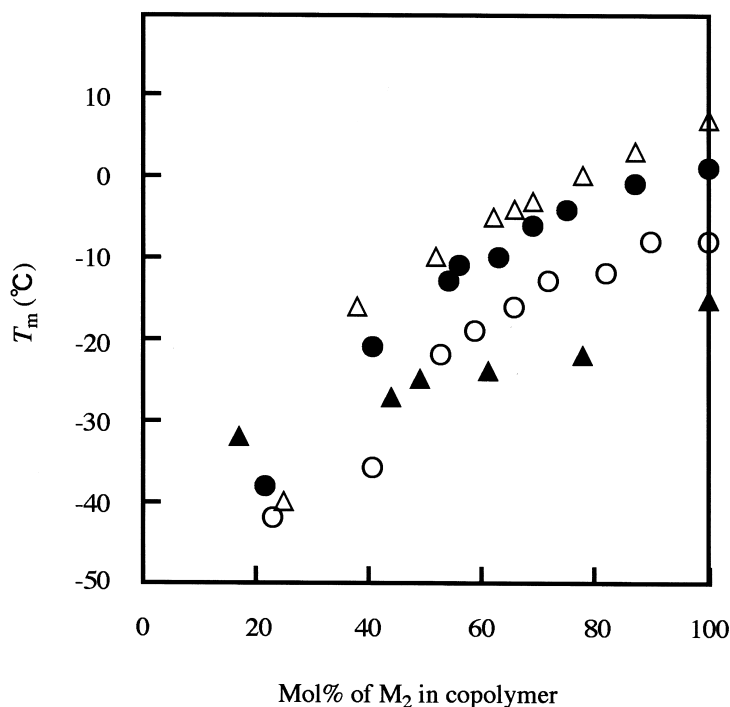
Polymer	$T_m$ , °C	$T_g$ , °C	$\Delta H_m$ , kJ/unit·mol	$N^a$
<b>P-1</b>	-8	112	10.88	3.5
<b>P-2</b>	1	103	15.10	4.9
<b>P-3</b>	7	96	18.45	6.0
Poly <b>N18</b> <sup>b</sup>	-15	132	6.02	2.0

<sup>a</sup>Number of crystallizing CH<sub>2</sub> groups calculated from  $\Delta H_m$ .

<sup>b</sup>Ref. 8.

alkyl side chains of the polymers crystallized in hexagonal form. Therefore, the number of crystallizing methylene groups per alkyl side chain ( $N$ ), as the indication of degree of crystallinity, was calculated from  $\Delta H_m$  on the basis of the heat of fusion of n-alkanes in hexagonal packing (3.08 kJ/CH<sub>2</sub> mol) [3]. Values of  $N$  are also listed in Table 3.

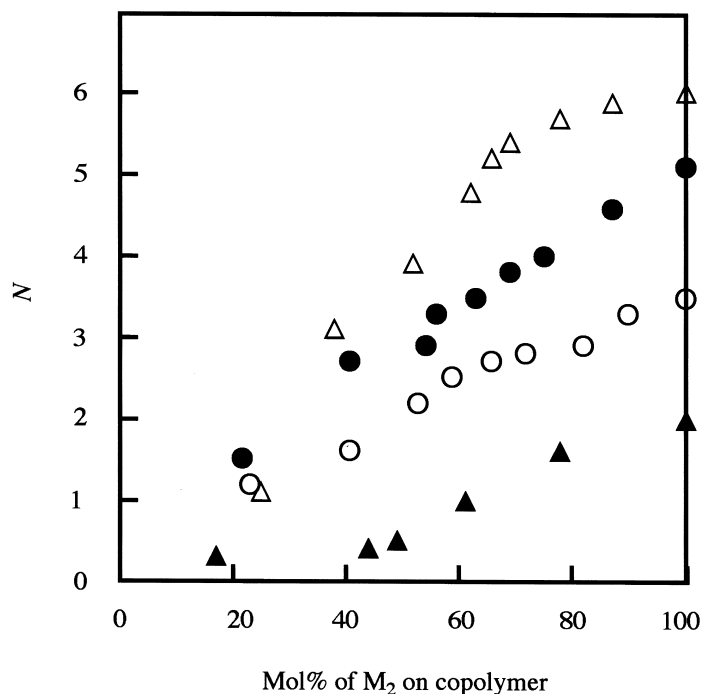
As can be seen in Table 3,  $T_m$  and  $N$  values of the present comb-like polymers were higher than those of Poly**N18**, and they increased with increasing the length of oxyethylene spacer. It was proved that the introduction of oxyethylene



**Figure 3.** Relationship between melting temperature of side chains  $T_m$  and copolymer composition. Copolymer types: poly(1-co-styrene) (O); poly(2-co-styrene) (●); poly(3-co-styrene) (Δ); poly(N18-co-styrene) (▲).

spacer between polymer backbone and long alkyl group raise the perfection of the crystals and degree of crystallinity. The increase in the crystallinity of side chains would be due to the rise in flexibility of alkyl side chain, caused by the insertion of flexible spacer, favorable to side chain packing. This is supported by the previous reports [2, 18-20] that crystallization of side chains in comb-like polymers depends remarkably on the flexibility of the main chain.

In addition,  $T_g$  and melting endothermic peaks were also observed in DSC curves of copolymers.  $T_g$  of the copolymers was almost kept constant at around  $100^\circ\text{C}$  independently of the composition of the copolymers. On the other hand,  $T_m$  and  $\Delta H_m$  depended on the copolymer composition.  $T_m$  and  $N$  calculated from  $\Delta H_m$  are plotted against the copolymer composition in Figures 3 and 4, respectively. Previous results for **N18** copolymers with styrene are also shown in these Figures.  $T_m$  and  $N$  values decreased with increasing the content of styrene as can be seen in Figures 3 and 4. This result indicated that the perfection of the crystals and degree of crystallinity of long alkyl side chains in the comb-like copolymers depend significantly on the concentration of the long alkyl side chain.  $T_m$  and  $N$  values of the present copolymers were higher than those of **N18** copolymers, and they increased with the length of oxyethylene spacer. This can be explained that the crystallinity



**Figure 4.** Relationship between the number of crystallizing  $\text{CH}_2$  groups in a side chain ( $N$ ) and copolymer composition. Symbols as in Figure 3.

of side chains in the comb-like copolymers is increased by the rise in flexibility of alkyl side chain caused by the insertion of flexible spacer similarly to the homopolymers as mentioned above.

## CONCLUSION

Isopropenyl-1,3,5-triazine monomers carrying a octadecyl group with oxyethylene spacer between the triazine ring and the alkyl group were synthesized from 2-amino-4-anilino-6-isopropenyl-1,3,5-triazine (**AAIT**). The numbers of oxyethylene unit were 1, 2, and 3. Homopolymers could be obtained by polymerization of these monomers in benzene using 2,2'-azobisisobutyronitrile. Copolymerization of these monomers with styrene was carried out and the monomer reactivity ratios were determined. The monomer reactivity was independent of the length of oxyethylene spacer. The long alkyl side chains in the resulting comb-like polymers could be crystallized below the glass transition temperature of polymers. The crystallinity of the side chains evaluated by differential scanning calorimetry increased by the introduction of flexible spacer and with increasing the length of oxyethylene spacer.

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