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## Journal of Macromolecular Science, Part A

Publication details, including instructions for authors and subscription information:

<http://www.informaworld.com/smpp/title~content=t713597274>

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Online publication date: 23 August 1999

**To cite this Article** Xu, Hongyao and Tang, Ben Zhong(1999) 'SYNTHESES AND OPTICAL PROPERTIES OF POLY(C<sub>60</sub>-CO-PHENYLACETYLENE)S', Journal of Macromolecular Science, Part A, 36: 9, 1197 – 1207

**To link to this Article:** DOI: 10.1081/MA-100101592

**URL:** <http://dx.doi.org/10.1081/MA-100101592>

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## SYNTHESES AND OPTICAL PROPERTIES OF POLY(C<sub>60</sub>-CO-PHENYLACETYLENE)S

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Key Words: C<sub>60</sub>, Polyacetylene, C<sub>60</sub>-Phenylacetylene Copolymer, Optical Limiting

### ABSTRACT

Copolymers of C<sub>60</sub> and phenylacetylene (PA) were synthesized by copolymerization reactions catalyzed by MoCl<sub>5</sub>Ph<sub>4</sub>Sn, WCl<sub>6</sub>Ph<sub>4</sub>Sn, [Rh(nbd)]Cl<sub>2</sub>, and [Rh(cod)]Cl<sub>2</sub> (where nbd = 2,5-norbornadiene, cod = 1,5-cyclooctadiene). Stirring a toluene solution of C<sub>60</sub>, phenylacetylene, and the catalyst under nitrogen at room temperature for 24 hours produced poly(C<sub>60</sub>-co-phenylacetylene)s in good yields. Varying the C<sub>60</sub> feed ratio changed the C<sub>60</sub> content of the copolymers from 0.58 to 13.7 wt%. All the copolymers are soluble in common solvents such as THF and chloroform, and the structures of the copolymers were characterized by spectroscopic methods. The C<sub>60</sub>-phenylacetylene copolymers effectively limited strong 532-nm laser pulses.

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## INTRODUCTION

The synthesis and isolation of gram quantities of  $C_{60}$  and the discovery of its novel materials properties including superconductivity, ferromagnetism, and optical nonlinearity have stimulated a flurry of research efforts in fullerene science. The poor tractability of  $C_{60}$ , however, greatly hampers the utilization of the properties for practical applications. Synthesis of fullerenated polymers has attracted much interest of polymer scientists because suitably designed fullerene polymers would be well processable and may exhibit novel properties that are superior to those of the parent forms, thus generating new fullerene-based specialty materials. Many different kinds of polymers containing  $C_{60}$ , including “pearl necklace”, “charm bracelet”, and “flagellene”, have been prepared [1-10]. The preparation of the polymers, however, sometimes requires nontrivial synthetic efforts in prefunctionalizing  $C_{60}$  and monomers/polymers, which in turn limit the scope of applicability of the methodology.

Our group has worked on the development of simple polymer reactions with general applicability for directly attaching  $C_{60}$  to preformed polymers [11-13]. Polyacetylene is a prototypical conjugated polymer and its doped form possesses high conductivity. High molecular weight substituted polyacetylenes have shown interesting properties such as photoconductivity, gas permeability, optical nonlinearity, and liquid crystallinity [14-21]. In this work, we tried to incorporate  $C_{60}$  into polyacetylene chains. Here, we report the synthesis and optical properties of poly( $C_{60}$ -*co*-phenylacetylene)s [ $P(C_{60}$ -PA)s].

## EXPERIMENTAL

### Materials

High-purity (99.95%)  $C_{60}$  was purchased from MER and kept in the dark before use. Phenylacetylene was purchased from Farchan, distilled from calcium hydride at reduced pressure, and stored in sealed ampules in a dark and cold place. 2,5-Norbornadiene rhodium(I) chloride dimer  $\{[Rh(nbd)Cl]_2\}$ , 1,5-cycloocta-diene rhodium(I) chloride dimer  $\{[Rh(cod)Cl]_2\}$ , tungsten(VI) chloride, tetraphenyltin (all from Aldrich), and molybdenum(V) chloride (Acros) were used as received without further purification. Toluene was purchased from Aldrich, dried over 4 Å molecular sieves, and distilled from sodium benzophenone ketyl immediately prior to use. Technical grade methanol was used to precipitate the polymer products.

### Polymerization

All the polymerization reactions were carried out under nitrogen. In a typical run, 33 mg of C<sub>60</sub>, 97 mg of WCl<sub>6</sub> (0.24 mmol), and 103 mg of Ph<sub>4</sub>Sn (0.24 mmol) were added into a prebaked 20-mL Schlenk polymerization tube under nitrogen. Freshly distilled toluene (5 mL) and 0.55 mL (5.0 mmol) of purified phenylacetylene were then injected into the above polymerization tube by a syringe under nitrogen at room temperature. After stirring at room temperature for 24 hours, the reaction was quenched with 5 mL of toluene containing a small amount of methanol, and the diluted reaction mixture was filtered by a cotton filter. The soluble filtrate was added dropwise into 400 mL of methanol under stirring to precipitate the polymer product. The product was redissolved in tetrahydrofuran (THF) and the resulting solution was centrifuged at 2 000 rpm for 16 minutes. The supernatant was added dropwise through a cotton filter into hexane (ca. 300 mL). The precipitate was collected and then dried under vacuum at 40°C to a constant weight. An orange red powder was obtained in 45 wt% yield, whose C<sub>60</sub> content was estimated to be 13.7 wt%.

### Instrumentation

The <sup>1</sup>H and <sup>13</sup>C NMR spectra were recorded on a Bruker ARX300 spectrometer using chloroform-*d* as solvent. Tetramethylsilane (TMS) or chloroform-*d* was used as internal reference for the NMR analysis. Chemical shifts are reported in parts per million (ppm). The FTIR spectra were measured on a Perkin-Elmer 16 PC spectrophotometer (KBr). The UV-Vis absorption and transmission spectra were recorded on a Milton Roy Spectronic 3000 Array spectrophotometer using a 1-cm square quartz cell. The molecular weights (MWs) of all the polymers were estimated by a Waters gel permeation chromatograph (GPC) using a Waters Associates liquid chromatograph equipped with a Waters 510 HPLC pump, a Rheodyne 7725I injector with a stand kit, a set of styragel columns (HT3, HT4, and HT6; molecular weight range: 10<sup>2</sup>-10<sup>7</sup> g/mol), a column temperature controller, a Waters 486 wavelength-tunable UV-Vis detector, a Waters 410 differential refractometer, and a system DMM/scanner with an 8-channel scanner option. All the polymer solutions were prepared in degassed THF (ca. 2 mg/mL) and filtered through 0.45-μm PTFE syringe-type filters before being injected into the GPC system. Degassed THF was used as eluent at a flow rate of 1.0 mL/min. The working wavelength of the UV detector in the GPC system was set at 254 nm. A set of 12 Waters monodisperse polystyrene standards were used for calibration.

### Optical Limiting Measurement

The optical limiting experiments were performed at 532 nm with 8-ns pulses from a frequency-doubled Q-switched Nd:YAG laser (Quanta Ray GCR-3) operating in a near-Gaussian transverse mode with a repetition rate of 10 Hz. The pulsed laser beam was focused into a 1-cm square quartz cell filled with a THF solution of P(C<sub>60</sub>-PA). The incident and transmitted energies were measured by an OPHIR detector (30-A-P-Diff-SH). Every point of the optical limiting data was the average of at least 15 laser shots. The detector was connected to a computer both before and after the optical limiting measurements were run, and the output stability of the laser equipment was double checked by taking a series of output data by the energy meter every 10 s for an extended period of time.

## RESULTS AND DISCUSSION

### Synthesis and Characterization

It is known that substituted alkynes and strained cycloalkenes can be polymerized by Mo and W catalysts [14]. C<sub>60</sub> is a strained cycloalkene and may undergo copolymerization reactions with alkynes via metathesis polymerization mechanisms. We thus polymerized phenylacetylene in the presence of C<sub>60</sub> with a C<sub>60</sub> feed ratio of 6.5 wt% using WCl<sub>6</sub>-Ph<sub>4</sub>Sn as catalyst. C<sub>60</sub> was found to be consumed in the polymerization reaction. To check whether C<sub>60</sub> has chemically incorporated into polyphenylacetylene chains, we isolated and purified the polymer product by repeated dissolution in THF, which is not a solvent of C<sub>60</sub> [22] but a solvent of polyphenylacetylene. After centrifugation, the supernatant was added dropwise through a filter into hexane, a solvent of C<sub>60</sub> [23] but a nonsolvent of polyphenylacetylene. The purification processes were repeated at least 3 times. No unreacted C<sub>60</sub> was recovered; in other words, all the C<sub>60</sub> had reacted. The P(C<sub>60</sub>-PA) was completely soluble in THF, indicating that there was no unreacted C<sub>60</sub> particles physically wrapped in the polymer product, which allows us to estimate the C<sub>60</sub> content of the copolymer on the basis of C<sub>60</sub> feed ratio. The C<sub>60</sub> content in the P(C<sub>60</sub>-PA) was 13.7 wt% (Table 1, no. 1), but the average C<sub>60</sub> number per PPA chain calculated on the *M<sub>n</sub>* data was only 0.68, that is, every 100 PPA chains were attached with 68 C<sub>60</sub> cages on the average.

We further investigated copolymerization behavior of C<sub>60</sub> and phenylacetylene using MoCl<sub>5</sub>-Ph<sub>4</sub>Sn, [Rh(cod)Cl]<sub>2</sub>, and [Rh(nbd)Cl]<sub>2</sub> as catalysts. C<sub>60</sub>

TABLE 1. Polymerization of Phenylacetylene (PA) in the Presence of C<sub>60</sub><sup>a</sup>

No.	C <sub>60</sub> /PA <sup>b</sup>	Catalyst <sup>c</sup> (mg)	Cocatalyst (mg)	Yield		C <sub>60</sub> content <sup>e</sup>	
	(wt %)			(wt %)	$M_w/10^{3d}$	$M_w/M_n^d$	(wt %)
1	6.5	WCl <sub>6</sub> (97)	Ph <sub>4</sub> Sn (104)	45	6	1.7	13.7
2	5.9	MoCl <sub>5</sub> (102)	Ph <sub>4</sub> Sn (102)	47	6	2.6	11.7
3	5.9	[Rh(nbd)Cl] <sub>2</sub> (84)		52	11	3.6	10.6
4	5.9	[Rh(cod)Cl] <sub>2</sub> (84)		62	16	1.9	8.9

<sup>a</sup> Carried out under nitrogen in toluene (5 mL) at room temperature for 24 h.

<sup>b</sup> Feed amount of PA: 0.55 mL (512 mg).

<sup>c</sup> Abbreviation: nbd = 2,5-norbornadiene, cod = 1,5-cyclooctadiene.

<sup>d</sup> Estimated by GPC on the basis of a polystyrene calibration.

<sup>e</sup> Estimated by mass ratio.

was also consumed and copolymerized with phenylacetylene in these polymerization reactions (Table 1). The Rh complexes generally produced polymers with higher molecular weights in higher yields.

We studied the effect of the C<sub>60</sub> feed amount on the copolymerization (Table 2). Varying the C<sub>60</sub> feed ratio from 0.6 to 6.5 wt%, the average amount of C<sub>60</sub> cages attached to PPA chains changed from 0.9 to 13.7 wt%. With increasing the feed amount of C<sub>60</sub>, the  $M_w$  and the yield of the P(C<sub>60</sub>-PA) generally decreased. The same phenomenon was found in [Rh(nbd)Cl]<sub>2</sub> catalyst system (Table 2, nos. 6 and 7). Thus, C<sub>60</sub> seems to play a role in hindering the propagation process of the PPA chain. The color of the P(C<sub>60</sub>-PA) varies from orange to orange-red with increasing the C<sub>60</sub> content, while the parent poly-phenylacetylene is yellow in color. The color change supports the incorporation of the C<sub>60</sub> cages into the polymer chains.

Figure 1 shows FTIR spectra of C<sub>60</sub>, polyphenylacetylene, and P(C<sub>60</sub>-PA). Although the spectra of the homopolymer and copolymer look similar, careful analysis provides some distinctions. A weak and broad characteristic absorption band at 526 cm<sup>-1</sup>, which is due to the C<sub>60</sub> moiety (cf. Figure 1A), was observed in the spectrum of the copolymer. The absorption band at 526 cm<sup>-1</sup> often survives in the functionalization of C<sub>60</sub> [24].

TABLE 2. Effect of C<sub>60</sub>/PA Feed Ratio on Polymerization of Phenylacetylene (PA)<sup>a</sup>

No.	C <sub>60</sub> /PA <sup>b</sup>		C <sub>60</sub> content <sup>d</sup>		
	(wt %)	Yield (wt %)	M <sub>w</sub> /10 <sup>3c</sup>	M <sub>w</sub> /M <sub>n</sub> <sup>c</sup>	(wt %)
<b>Catalyzed by WCl<sub>6</sub> (97 mg) and Ph<sub>4</sub>Sn (103 mg)</b>					
1	0	92	16	1.9	0
2	0.6	93	16	2.1	0.9
3	2.3	70	10	2.2	2.5
4	4.4	86	13	2.3	4.9
5	6.5	45	6	1.7	13.7
<b>Catalyzed by [Rh(nbd)Cl]<sub>2</sub> (4 mg)<sup>e</sup></b>					
6	0	100	97	4.09	0
7	5.9	68	15	1.94	8.7

<sup>a</sup> Carried out under nitrogen in toluene (5 mL) at room temperature for 24 h.

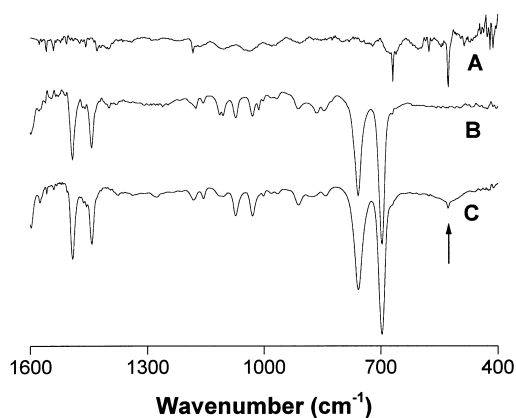
<sup>b</sup> Feed amount of PA: 0.55 mL (512 mg).

<sup>c</sup> Estimated by GPC on the basis of a polystyrene calibration.

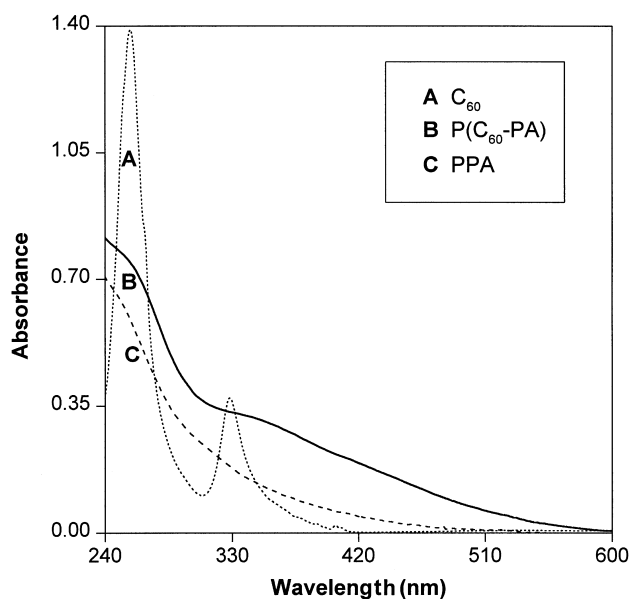
<sup>d</sup> Estimated by mass ratio.

<sup>e</sup> Abbreviation: nbd = 2,5-norbornadiene.

Additional evidence supporting the incorporation of the fullerene units into the polymer chains was obtained from the UV-Vis spectroscopic analysis. C<sub>60</sub> absorbs at 255.8 and 330 nm and its absorption edge locates at 412 nm, while PPA absorption is a steadily decreasing curve with an absorption edge at 502 nm (Figure 2). The electronic absorption of P(C<sub>60</sub>-PA) shows two shoulder peaks at 260 and 326 nm with the absorption edge well extending to 600 nm. The absorption at 326 nm is beyond doubt from the C<sub>60</sub> moiety in the copolymer and the red-shift in the absorption edge indicates the enhancement in the  $\pi$ -conjugation of the copolymer.



**Figure 1.** IR spectra of (A) C<sub>60</sub>, (B) polyphenylacetylene (sample from Table 2, no. 1), and (c) poly(C<sub>60</sub>-co-phenylacetylene) (sample from Table 2, no. 4).



**Figure 2.** UV spectra of (A) C<sub>60</sub> in hexane (0.011 mg/mL), (B) poly(C<sub>60</sub>-co-phenylacetylene) in THF (0.012 mg/mL; sample from Table 2, no. 4), and (C) polyphenylacetylene in THF (0.012 mg/mL; sample from Table 2, no. 1).

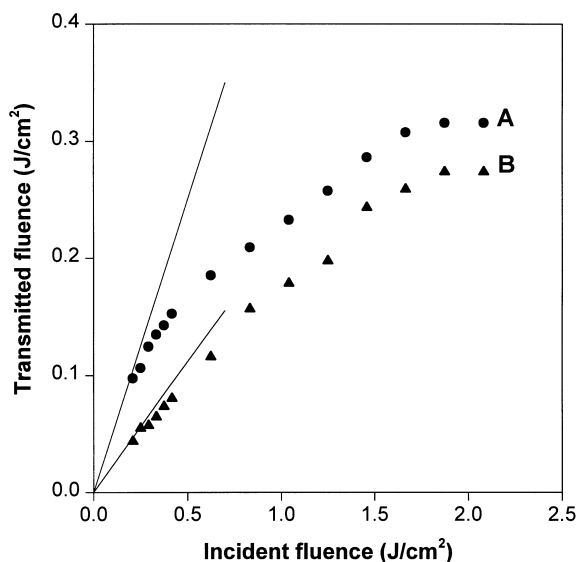


The  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra of the chloroform solutions of the  $\text{P}(\text{C}_{60}\text{-PA})$ s were similar to those of the parent PPA. It may be due to the relatively small number of the  $\text{C}_{60}$  cages attached to the polymer chains [5, 11, 25-27].

### Optical Property

Development of optical limiters is a topic of great current interest because of the rapid advancement in the development of ultrafast high-power laser and the growth in space exploration [28, 29].  $\text{C}_{60}$  is an excellent optical limiter [30], but its poor tractability has been an obstacle in finding practical applications for the limiting property [22, 31-33].

The  $\text{P}(\text{C}_{60}\text{-PA})$ s have better solubility than the parent  $\text{C}_{60}$ , which enables the preparation of high-concentration solutions. The polymer solutions were shot with 532-nm laser pulses. The incident and transmitted laser-pulse energies were measured and the plot of transmitted fluence versus the incident fluence is shown in Figure 3. In the low fluence, a THF solution (3.0 mg/mL) of  $\text{P}(\text{C}_{60}\text{-PA})$  with a  $\text{C}_{60}$  content of 4.9 wt% exhibited a high linearity transmittance (60%). In the high fluence region, the  $\text{P}(\text{C}_{60}\text{-PA})$  solution strongly limited the intense laser



**Figure 3.** Optical limiting responses to 8-ns, 532-nm optical pulses, of THF solutions of poly( $\text{C}_{60}\text{-co-phenylacetylene}$ ) sample from Table 2, no. 4); concentration (mg/mL)/linear transmittance (%): (A) 3.00/60, (B) 4.00/30.

pulses. The transmitted fluence started to deviate from linearity at incident fluence of ca. 0.25 and 0.23 J/cm<sup>2</sup> for the solutions with linear transmittance of 60% and 30%, respectively. The transmittance of the solutions was unchanged in the region of low incident fluence but rapidly decreased when subjected to high-intensity radiation. This clearly shows that the absorption coefficient of the solutions increased with increasing the incident fluence. The optical limiting property is contributable to the C<sub>60</sub> moiety in the P(C<sub>60</sub>-PA) solutions because the parent PPA does not show optical limiting property at the same concentrations [21]. The optical limiting behavior further confirms that the C<sub>60</sub> cages are attached to the PPA chains. As can be seen from Figure 3, the optical limiting property is affected by concentration. When the concentration of solution changed from 3.0 to 4.0 mg/mL, the limiting power was enhanced.

## CONCLUSION

In this study, direct copolymerization of C<sub>60</sub> and phenylacetylene has been achieved. Stirring a toluene solution of C<sub>60</sub> and PA in the presence of the metal catalysts at room temperature steadily attaches C<sub>60</sub> to PPA chains. The P(C<sub>60</sub>-PA)s are readily soluble and its solutions effectively limit strong laser pulses. Because of  $\pi$  electronic interaction between C<sub>60</sub> cages and polyacetylene chains, the copolymers may possess interesting materials properties. This work thus not only provides a versatile synthetic tool for preparing processible fullerene-functionalized polyacetylenes but also offer attractive opportunities for exploring new fullerene-based specialty materials with unique optical and electronic properties.

## ACKNOWLEDGMENT

This work was in part supported by the Hong Kong Research Grants Council (HKUST 6062/98P and DAG 96/97.SC11).

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Received February 15, 1999

Revision received April 5, 1999