## **Supporting Information**

## Photohardenable Pressure-Sensitive Adhesives using Poly(methyl methacrylate) containing Liquid Crystal Plasticizers

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## Measurement of tack strength of PP<sub>x</sub>/PMMA mixtures.

Tack strength was measured by a probe tack test. Probe tack test was conducted in the rheometer using an aluminum probe under the following conditions: temperature was 25 °C, the approaching and debonding speeds were 0.5 mm/s, the contact pressure was  $1 \pm 0.2$  N/cm<sup>2</sup>, and the contact time was 1 s. To investigate the tackiness of photoplasticized samples, the tack strength of the samples pre-irradiated with UV light (wavelength: 365 nm; intensity: 30 mW/cm<sup>2</sup>) for 30 min were measured under UV-light irradiation.

## **Supplementary figures**



**Figure S1.** UV–vis absorption spectral changes of BMAB in THF before UV-light irradiation (solid line) and after UV-light (dotted line; wavelength: 365 nm; intensity: 30 mW/cm<sup>2</sup>) irradiation, and after Vis-light (dot-dashed line; wavelength: 435 nm; intensity: 30 mW/cm<sup>2</sup>) irradiation following the UV-light irradiation.



**Figure S2.** The XRD patterns of (A) PP<sub>5</sub>, (B) PP<sub>50</sub>, and (C) PP<sub>100</sub> after UV-light irradiation at 25 °C.



**Figure S3**. The DSC curves of (A) PP<sub>5</sub>, (B) PP<sub>50</sub>, and (C) PP<sub>100</sub> before (a) and during (b) UV-light irradiation.



**Figure S4.** Polarized optical micrographs of (A) PP<sub>50</sub>/PMMA and (B) PP<sub>100</sub>/PMMA before UVlight irradiation at 25 °C. P and A denote polarizer and analyzer, respectively.



**Figure S5.** Temperature dependences of the viscoelastic parameters of  $PP_x/PMMA$  mixtures before (black lines) and under (red lines) UV-light irradiation (wavelength: 365 nm; intensity: 30 mW/cm<sup>2</sup>). (A) and (B) show the *G*' (solid lines) and *G*'' (dashed lines) values of PP<sub>5</sub>/PMMA and PP<sub>100</sub>/PMMA, respectively. (C) and (D) show the tan $\delta$  values of PP<sub>5</sub>/PMMA and PP<sub>100</sub>/PMMA, respectively. The viscoelastic parameters were measured at a frequency of 1 Hz and a strain of 0.1% upon heating at 1 °C/min.



**Figure S6.** The viscoelastic parameters of (A) PP<sub>5</sub>/PMMA and (B) PP<sub>100</sub>/PMMA at 25 °C upon the alternate irradiation of UV (wavelength: 365 nm; intensity: 30 mW/cm<sup>2</sup>) and Vis (wavelength: 435 nm; intensity: 30 mW/cm<sup>2</sup>) light; solid line: G'; dashed line: G''.

**Table S1.** Tack strengths of PP<sub>5</sub>/PMMA, PP<sub>50</sub>/PMMA, and PP<sub>100</sub>/PMMA at 25 °C under UV-light irradiation (wavelength: 365 nm; intensity: 30 mW/cm<sup>2</sup>).

Sample	Tack strength under UV-light irradiation (mN/mm <sup>2</sup> )
PP <sub>5</sub> /PMMA	$7.3 \pm 1.3$
PP <sub>50</sub> /PMMA	$3.0\pm0.4$
PP <sub>100</sub> /PMMA	$5.6 \pm 1.4$



**Figure S7.** The stress-strain curves of PP<sub>5</sub>/PMMA (dotted line), PP<sub>50</sub>/PMMA (solid line), and PP<sub>100</sub>/PMMA (dot-dashed line) at 25 °C.



**Figure S8.** The XRD pattern of the PP<sub>50</sub>/PMMA film at 25 °C with polyimide sheets. The peak labeled as "X" originates from the polyimide sheet.



**Figure S9**. Typical stress-strain curves obtained in a lap-shear test of PP<sub>50</sub>/PMMA sample specimens prepared in (A) photochemical and (B) thermal manners.