Supporting Information

Distinctive Field-Effect Transistors and Ternary Inverters Using Cross-Type WSe$_2$/MoS$_2$ Heterojunctions Treated with Polymer Acid

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Figure S1. (a) AFM image of poly(methyl methacrylate-co-methacrylic acid) (PMMA-co-PMAA) film with a profile line (white line). (b) Cross-sectional profile of the AFM image along the white line in (a).

The thickness of poly (methyl methacrylate-co-methacrylic acid) (PMMA-co-PMAA) film was measured by using atomic force microscope (AFM; Nano-Focus Ltd., Albatross). Figures S1(a) and (b) show the AFM image of PMMA-co-PMAA film with a profile line (white line) and the cross-sectional profile of the image, respectively. The PMMA-co-PMAA in ethyl lactate was spin-coated on the SiO$_2$/Si substrate at 4500 rpm for 1 min, then baked at 150 °C for 1 min. To measure the thickness of the fabricated PMMA-co-PMAA film, the side square-steps with 15 μm were made by using e-beam lithography, as shown in Fig. S1(a). The thickness of PMMA-co-PMAA film was measured to be approximately 305 nm, as shown in Fig. S1(b).
Figure S2. Deconvolution of LCM PL peaks for (a) pristine WSe\textsubscript{2} and (b) WSe\textsubscript{2} with PMMA-co-PMAA. Deconvolution of LCM PL peaks for (c) pristine MoS\textsubscript{2} and (d) MoS\textsubscript{2} with PMMA-co-PMAA.

Figure S2(a) shows the deconvolution of LCM PL peak for pristine WSe\textsubscript{2}. The LCM PL peak for pristine WSe\textsubscript{2} was deconvoluted into two peaks centered at 741 and 752 nm, which originated from neutral exciton and positive trion, respectively. The percentage of neutral exciton and positive trion for the pristine WSe\textsubscript{2} was estimated to be approximately 70 \% and 30 \%, respectively. After PMMA-co-PMAA treatment, the percentage of neutral exciton (centered at 743 nm) and positive...
trion (centered at 751 nm) of the WSe$_2$ was estimated to be approximately 46 % and 54 %, respectively, resulting in charge transfer effect.

Figure S2(c) shows the deconvolution of LCM PL peak for pristine MoS$_2$. The LCM PL peak for pristine MoS$_2$ was deconvoluted into three peaks centered at 606, 653, and 667 nm, which originated from $B$-exciton, neutral exciton, and negative trion, respectively. The percentage of neutral exciton and negative trion for the pristine MoS$_2$ was estimated to be approximately 40% and 45%, respectively. After PMMA-co-PMAA treatment, the percentage of neutral exciton (centered at 655 nm) and negative trion (centered at 669 nm) of the MoS$_2$ was estimated to be approximately 25% and 60%, respectively, resulting in charge transfer effect.
Figure S3. XPS spectra of S 2p for pristine MoS$_2$ (black curve) and MoS$_2$ with PMMA-co-PMAA (red curve).

Figure S3 shows the XPS spectra of S 2p for MoS$_2$ before and after PMMA-co-PMAA treatment. The XPS peaks of S 2p$_{3/2}$ and S 2p$_{1/2}$ for the pristine MoS$_2$ were observed at 162.18 and 163.38 eV. After PMMA-co-PMAA treatment, both the XPS peaks were up-shifted to higher binding energies ($\Delta_{BE} = 0.40$ eV), which indicated that the Fermi level was up-shifted closer to conduction band by n-type doping.
Figure S4. (a) XRD patterns of pristine MoS$_2$ (black curve) and MoS$_2$ with PMMA-co-PMAA (red curve). (b) XRD patterns of WSe$_2$ before (black curve) and after (red curve) PMMA-co-PMAA treatment.

Table S1. Comparison of full width at half maximum (FWHM) of XRD peaks of MoS$_2$ and WSe$_2$ before and after PMMA-co-PMAA treatment

<table>
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<tr>
<th>Material</th>
<th>Miller index</th>
<th>FWHM (degree)</th>
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<tbody>
<tr>
<td></td>
<td></td>
<td>Before treatment</td>
</tr>
<tr>
<td>MoS$_2$</td>
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<td>0.14</td>
</tr>
<tr>
<td></td>
<td>(104)</td>
<td>0.22</td>
</tr>
<tr>
<td></td>
<td>(008)</td>
<td>0.22</td>
</tr>
<tr>
<td>WSe$_2$</td>
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<td>0.12</td>
</tr>
<tr>
<td></td>
<td>(006)</td>
<td>0.088</td>
</tr>
<tr>
<td></td>
<td>(008)</td>
<td>0.088</td>
</tr>
<tr>
<td></td>
<td>(0010)</td>
<td>0.088</td>
</tr>
</tbody>
</table>
Figure S4 (a) shows the XRD patterns of MoS$_2$ before and after PMMA-co-PMAA treatment. The XRD peaks of the pristine MoS$_2$ were observed at (002), (104) and (008) planes.$^{1-3}$ After PMMA-co-PMAA treatment, XRD peaks were observed at the same planes and the full-width at half maximum (FWHM) of the observed XRD peaks as listed in Table S1 were not changed in comparison to FWHM of XRD peaks for the pristine MoS$_2$. Figure S4 (b) shows the XRD patterns of WSe$_2$ before and after PMMA-co-PMAA treatment. The XRD peaks of the pristine WSe$_2$ were observed at (002), (006), (008), and (0010) planes.$^4$ With PMMA-co-PMAA treatment, XRD peaks of WSe$_2$ were the same, however the XRD peak intensity at (0010) plane decreased.
Figure S5. FT-IR spectra of pristine PMMA-co-PMAA (black line), MoS$_2$/PMMA-co-PMAA (red line), and WSe$_2$/PMMA-co-PMAA (blue line).

Figure S5 shows the FT-IR spectra of PMMA-co-PMAA, MoS$_2$/PMMA-co-PMAA, and WSe$_2$/PMMA-co-PMAA. The FT-IR characteristics peaks for C-O, -CH$_3$, and C=O modes from PMMA-co-PMAA were observed at 1144, 1440, and 1725 cm$^{-1}$, respectively.$^5$ The FT-IR peaks from those modes were not much changed by the hybrid with MoS$_2$ or WSe$_2$. 
Figure S6. Comparison of transfer characteristic curves on logarithmic scale for pristine (black curve) and PMMA-co-PMAA treated (red curve) (a) MoS$_2$ and (b) WSe$_2$ ($V_D = 0.1$ V) with forward and backward biases. The solid and open markers represent the forward and backward biases, respectively. FET: field-effect transistor; $V_D$: source-drain bias; $V_G$: gate bias; $I_D$: source-drain current.

Figure S6(a) shows the transfer characteristic curves of the pristine (black curve) and PMMA-co-PMAA treated (red markers) MoS$_2$-based field-effect transistor (FET) with forward (solid markers) and backward (open markers) biases. The hysteresis of the transfer characteristics of the FET was drastically reduced after PMMA-co-PMAA treatment. Figure S6(b) shows the transfer characteristic curves of the pristine (black markers) and PMMA-co-PMAA treated (red markers) WSe$_2$-based FET. The hysteresis of the transfer characteristic curve of the WSe$_2$-based FET was also reduced after PMMA-co-PMAA treatment due to surface passivation by the carboxyl acid groups in PMMA-co-PMAA. PMMA-co-PMAA treatment helps to enhance the doping, causing the source-drain current ($I_D$) of the MoS$_2$- and WSe$_2$-based FETs to increase in the specific regions.
of gate bias \( (V_G) \), and the threshold voltage \( (V_{th}) \) of MoS\(_2\) and the WSe\(_2\) FETs were negatively and positively shifted, respectively.
Figure S7. Output characteristic curves ($I_D$ vs. $V_D$) of the cross-type WSe$_2$/MoS$_2$-based FET with PMMA-co-PMAA treatment at (a) $V_G = 0$ V and (b) $V_G = -20$, $-30$, and $-35$ V.

The output characteristics ($I_D$ vs. $V_D$) of the cross-type WSe$_2$/MoS$_2$-based FET with PMMA-co-PMAA treatment are shown in Figs. S7(a) and (b). The rectifying behavior was clearly observed as a conventional $p$-$n$ junction diode. The output characteristics of the cross-type WSe$_2$/MoS$_2$-based FET were controlled by the $V_G$. 
**Figure S8.** (a) Schematic illustration of ternary inverter employing cross-type WSe$_2$/MoS$_2$-based FET subjected to PMMA-co-PMAA treatment. The W2, W1, and M1 electrodes were used for the driving voltage ($V_{dd}$), output voltage ($V_{out}$), and ground (GND), respectively. The device is the same that in Figs. 4–6; however, the electrodes for the $V_{dd}$ and $V_{out}$ were changed. (b) Transfer characteristic curves on logarithmic scale for pristine WSe$_2$/MoS$_2$-based FETs at source-drain bias ($V_D$) = 4, 6, and 8 V. (c) Transfer characteristic curves on logarithmic scale for PMMA-co-PMAA treated WSe$_2$/MoS$_2$-based FETs at $V_D$ = 4, 6, and 8 V. (d) Comparison of $V_{out}$ vs. input voltage ($V_{in}$) characteristic curves for ternary inverter employing cross-type WSe$_2$/MoS$_2$-based FET before (black markers) and after (red markers) PMMA-co-PMAA treatment at $V_{dd} = 6$ V (open markers) and 8 V (solid markers). (e) Ternary inverter characteristics (red markers) using the cross-type WSe$_2$/MoS$_2$-based FET with PMMA-co-PMAA treatment at $V_{dd} = 8$ V. Transfer characteristic curves using cross-type WSe$_2$/MoS$_2$-based FET with PMMA-co-PMAA treatment at $V_D = 8$ V.
(blue markers) and WSe$_2$-based FET with PMMA-co-PMAA treatment at $V_D = 0.1$ V (green markers). $V_G$: gate bias; $I_D$: source-drain current.

Figure S8(a) presents a schematic illustration of the ternary inverter employing the cross-type WSe$_2$/MoS$_2$-based FETs subjected to PMMA-co-PMAA treatment. This device is the same FET used for Figs. 5–7; however, the electrodes for the $V_{dd}$ and $V_{out}$ were changed. Figure S8(b) shows the transfer characteristic curves of the cross-type WSe$_2$/MoS$_2$-based FET with various source-drain biases ($V_D = 4$, 6, and 8 V) before PMMA-co-PMAA treatment. The transfer characteristic curves of the pristine WSe$_2$/MoS$_2$-based FET were obtained by using the W2 (for $V_D$) and M1 (for GND) electrodes, as shown in Fig. S8(a). The AAT and NDT characteristics of the cross-type WSe$_2$/MoS$_2$-based FET varied with different source-drain biases. At $V_D = 8$ V, the local maximum and valley state of the $I_D$ were measured to be $1.4 \times 10^{-9}$ A at $V_G = -61$ V and $2.5 \times 10^{-10}$ at $V_G = -55$ V, respectively. Figure S8(c) shows the transfer characteristic curves of the cross-type WSe$_2$/MoS$_2$ FET subjected to PMMA-co-PMAA treatment. Anti-ambipolar transistor (AAT) characteristics were clearly observed in the range of $-70$ V $\leq V_G \leq -40$ V at $V_D = 8$ V, where this range is much wider than that in which the pristine WSe$_2$/MoS$_2$-based FET exhibited AAT characteristics. The local maximum (peak) and valley state of the $I_D$ were measured to be $1.3 \times 10^{-9}$ A at $V_G = -65$ V and $1.5 \times 10^{-11}$ A at $V_G = -39$ V. The peak-to-valley current ratios of the pristine and PMMA-co-PMAA treated WSe$_2$/MoS$_2$ FET were estimated to be approximately 6 and 100, respectively. Figure S8(d) shows characteristic curves of the ternary inverter employing the WSe$_2$/MoS$_2$-based FET with various $V_{dd}$. Compared with the case in Fig. 7, the electrodes used for $V_{dd}$ and $V_{out}$ were changed from W1 and W2 to W2 and W1, respectively. After PMMA-co-PMAA treatment, stable and distinguishable logic states “1”, “1/2”, and “0” were realized for the cross-
type WSe$_2$/MoS$_2$-based FET. Figure S8(e) shows the characteristic curve (red markers) for the ternary inverter ($V_{dd} = 8$ V) and the transfer characteristic curves of the WSe$_2$/MoS$_2$-based FET ($V_D = 8$ V, blue markers) and the WSe$_2$-based FET ($V_D = 0.1$ V, green markers) after PMMA-co-PMAA treatment. The characteristics and operation mechanism of the ternary inverters from different batches were similar to those presented in Figs. 7 and 8.
**Figure S9.** (a) Transfer characteristic curves of WSe$_2$-based (blue markers) and WSe$_2$/MoS$_2$-based (black markers) FETs subjected to PMMA-co-PMAA treatment. (b) Transfer characteristic curves of pristine MoS$_2$-based FET (black curve) and MoS$_2$-based FET hybridized with $p$-type WSe$_2$ (red curve). $V_G$: gate bias; $I_D$: source-drain current.

Figure S9(a) shows the transfer characteristic curves of the effective $p$-type WSe$_2$-based (blue markers) and WSe$_2$/MoS$_2$-based (black markers) FETs subjected to PMMA-co-PMAA treatment. The results show that the slopes of the transfer characteristic curves with the red-dotted fitting lines are close to each other. This suggests that the resistance ratio of the effective $p$-type WSe$_2$-based FET was comparable to that of the WSe$_2$/MoS$_2$-based FET in the range of $-35 \leq V_G \leq -25$. Figure S9(b) shows the transfer characteristic curves of the pristine MoS$_2$-based FET (black curve) and the FET of MoS$_2$ overlapped with $p$-type WSe$_2$ (red curve). The $I_D$ and $V_{th}$ values of the FET of MoS$_2$ overlapped with $p$-type WSe$_2$ were clearly different from those of the pristine MoS$_2$-based FET.
References


