Supporting Information

Actin Cytoskeleton-Disrupting and Magnetic Field-Responsive Multivalent Supramolecular Assemblies for Efficient Cancer Therapy

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Figure S1. Synthetic routes of MNP-ABP-Ada (a) and HACD (b).
Figure S2. Characterization of MNP. (a) TEM image; (b) X-ray diffraction spectrum.

Figure S3. HPLC (a) and MS (b) analysis of the synthesized ABP peptide.
Figure S4. Magnified TEM image of MNP-APTES (a) and MNP-ABP-Ada (b). The low-contrast region (indicated by the red arrows) at the boundary of nanoaggregate showed the chemical coating of ABP-Ada onto the surface of MNPs.

Figure S5. Rapid assembly of MNP-ABP-Ada-HACD along with the direction of an artificial magnetic field (1.8 mT).

Figure S6. Inhibition of ABP addition on targeting of MNP-ABP-Ada-HACD to actin cytoskeleton.
Figure S7. Effect of MNP-ABP-Ada and MNP-ABP-Ada•HACD on cell death of non-cancerous cells. (a) Cell death assay (evaluated by propidium iodide staining) of NIH3T3 cells treated with MNP-ABP-Ada (50 mg/L) or MNP-ABP-Ada•HACD (50 mg/L plus 50 mg/L) assemblies in the absence of an AMF (−AMF) or in the presence of an AMF (+AMF). (b) Cell death assay of 293T cells. There is no significant difference between the groups ($P < 0.05$). Error bars indicate ± standard derivations.
Figure S8. Different uptake contents of MNP-ABP-Ada and MNP-ABP-Ada+HACD in tumor cells (A549) and non-cancerous cells (NIH3T3, 293T) after 24 h of co-incubation. The cells were washed three time by PBS buffer, and fluorescence intensity of the cells was measured by fluorescence microplate reader. * Indicates significant difference between the groups ($P < 0.05$). Error bars indicate ± standard derivations.
Figure S9. Effect of MNPs, MNP-MitP<sub>HACD</sub> and MNP-ABP-Ada<sub>HACD</sub> on actin cytoskeleton distribution of the A549 tumor cells under AMF treatment. Note that only MNP-ABP-Ada<sub>HACD</sub> assemblies have a great impact on actin cytoskeleton.
**Figure S10.** Effect of MNPs, MNP-MitP⊂HACD and MNP-ABP-Ada⊂HACD on cell death in the A549 tumor cells. * Indicates significant between-group difference ($P < 0.05$). The “ns” indicates that there is no significant difference between the groups. Error bars indicate ± standard derivations.
Figure S11. Temperature change of MNP-ABP-Ada and MNP-ABP-Ada$\subset$HACD in the absence of an AMF (−AMF) or in the presence of an AMF (+AMF). The solutions of MNP-ABP-Ada (50 mg/L) and MNP-ABP-Ada$\subset$HACD (50 mg/L plus 50 mg/L) were treated with AMF or not for indicated times, and then the temperature was determined by an infrared (IR) thermal imaging camera (Xintest, China). Error bars indicate ± standard derivations.
Figure S12. Iron contents in tumor tissues and representative organs in the MNP-ABP-Ada·HACD-treated mice. Iron assays were performed after 24 h of MNP-ABP-Ada·HACD injection. * Indicates significant between-group difference (P < 0.05). Error bars indicate ± standard derivations.

Figure S13. Body weight of the tumor-burden mice treated by MNP-ABP-Ada or MNP-ABP-Ada·HACD with the aid of AMF after 12 days. Note that there is no significant difference between the groups (P < 0.05).
Figure S14. Quantification of fluorescence intensity of FITC-tagged MNP-ABP-Ada and MNP-ABP-Ada-HACD bound to the A549-RFP tumor cells (a) and the non-cancerous NIH3T3 (b) and 293T cells (c) after 2 h of co-incubation. The remarkably increased relative fluorescence intensity of the assemblies on the tumor cells (evaluated by the Image J software) indicates that the assemblies could strongly bound to the tumor cells rather than the non-cancerous cells. * Indicates significant between-group difference ($P < 0.05$). Error bars indicate ± standard derivations.
**Table S1.** Statistical analysis of co-localization of Rho B-Pha and FITC-tagged MNP-ABP-Ada or MNP-ABP-Ada-\(\subset\)HACD in A549 cells (Figure 3a).

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<thead>
<tr>
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<th>Pearson's correlation coefficient (PCC)</th>
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<tr>
<td>MNP-ABP-Ada</td>
<td>0.656</td>
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<tr>
<td>MNP-ABP-Ada-(\subset)HACD</td>
<td>0.831</td>
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