Supporting Information

# Smart Magnetic Nanoplatform for Synergistic Anticancer Therapy by Maneuvering the Mussel-inspired Functional Magnetic Nanoparticles for pH Responsive Anticancer Drug Delivery and Hyperthermia

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#### Characterization of p(HEMA-co-DMA) abbreviated as (HEDO)

The structure of the HEDO particles was confirmed via 1H-NMR. Deuterated dimethyl sulfoxide (DMSO-d6) was used to dissolve p(HEMA-co-DMA), which results in a sharp peak at 2.5 ppm.

## XRD spectra of Fe<sub>3</sub>O<sub>4</sub> & HEDO-Fe<sub>3</sub>O<sub>4</sub>

The powder X-ray diffraction (XRD) pattern of the as-prepared Fe<sub>3</sub>O<sub>4</sub> and HEDO-Fe<sub>3</sub>O<sub>4</sub> are shown in Figure S2. Diffraction peaks appeared at  $2\theta = 18.2^{\circ}$  (111), 30.1° (220), 35.5° (311), 43.1° (400), 53.4° (422), 56.9° (511), 62.5° (440) and 73.9° (533), which is in good agreement with the characteristic peaks of standard magnetite crystal with a cubic spinel structure (PDF-00-019-0629) <sup>1</sup>. The line broadening of the XRD peaks can be a result of the small size of the particles. For HEDO-Fe<sub>3</sub>O<sub>4</sub>, all characteristic peaks were the same as those for Fe3O4 nanoparticles along with a broad peak at around  $2\theta=24^{\circ}$ . This peak may contribute to the typical peak of the carbon content in the polymer <sup>2</sup>.

## DLS of Fe<sub>3</sub>O<sub>4</sub> & HEDO-Fe<sub>3</sub>O<sub>4</sub>

The size distribution of the samples was studied using a very low concentration of Fe3O4 and HEDO-Fe<sub>3</sub>O<sub>4</sub> with a DLS instrument (Brookhaven Instruments Corporation). The mean diameters of Fe3O4 and HEDO-Fe<sub>3</sub>O<sub>4</sub> were measured as 13.3 nm and 18.9 nm, respectively. This result clearly indicates the existence of a thin layer of HEDO on the Fe<sub>3</sub>O<sub>4</sub> nanoparticles.

#### **UV-VIS photospectroscopy of HEDO, BTZ and HEDO-BTZ**

The availability of the catechol hydroxyl groups in HEDO after mixing with BTZ were observed via UV light absorption at 280 nm. As shown in the figure, a pure copolymer solution in DMF shows a peak at 280 nm due to catechol hydroxyl groups<sup>3</sup>, and a pure drug solution in DMF shows a peak at 270 nm due to the aromatic unit of the drug. Upon mixing the two

solutions, the peaks are affected, and after 24 h of mixing, both peak intensities significantly decrease due to the complexation of the drug to the catechol groups. As seen in the spectrum, the 280 nm peak is still detectable at this time, confirming the existance of the catechol hydroxyl groups in HEDO after mixing with BTZ. Furthermore the absence of an oxidation peak at 300 nm confirms that the catechol groups are not in their oxidized forms <sup>4</sup>. It should be mentioned that the presence of unoxidized and non-complexed catechol groups is essential for the ligand exchange processes to take place.



Figure S1. The 1H-NMR spectra of HEDO.



**Figure S2.** XRD spectra of Fe3O4 & HEDO-Fe<sub>3</sub>O<sub>4</sub>, indicating the all characteristic peaks of standard magnetite.



Figure S3. DLS showing the size distribution of A) Fe3O4 and B) HEDO-Fe<sub>3</sub>O<sub>4</sub>



**Figure S4.** UV-VIS photospectroscopy to prove the availability of catechol hydroxyl groups in HEDO after mixing with BTZ.

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