

## Supporting Information

### **Tumor Microenvironment Responsive FePt/MoS<sub>2</sub> Nanocomposites with Chemotherapy and Photothermal for Enhancing Cancer Immunotherapy Therapy**

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### Calculation of the photothermal conversion efficiency

The photothermal conversion efficiency of FePt/MoS<sub>2</sub> NPs were determined according to the previous report method.<sup>1,2</sup> The detailed calculation was given as following: 1 mL of FePt/MoS<sub>2</sub> NPs aqueous dispersion with different concentrations ([Fe]=100, 80, 50 and 20 µg mL<sup>-1</sup>) were exposed to 808 nm NIR laser for 10 min, and then the laser was shut off to allow the system to cool down to room temperature.

Then, the photothermal conversion efficiency ( $\eta$ ) value was calculated according to the equation 1:

$$\eta = \frac{Q_{in,mater}}{I(1-10^{-A_{808}})} \quad (1)$$

Where I is the laser power. A<sub>808</sub> is the absorbance of the FePt/MoS<sub>2</sub> NPs at the wavelength of 808 nm. Where  $Q_{in,mater}$  is the heat generation from FePt/MoS<sub>2</sub> NPs. During the photothermal heating process, total energy balance of this system as following equation 2:

$$mC_p \frac{dT}{dt} = Q_{in,mater} + Q_{in,sol} - Q_{out} \quad (2)$$

Where m and C<sub>p</sub> are the mass and heat capacity,  $Q_{in,sol}$  is the heat generation from water.  $Q_{out}$  is the energy loss to the surroundings. And the theory of heat transfer was calculated as following equation 3:

$$Q_{out} = hA(T - T_{surr}) \quad (3)$$

Where h is the heat transfer coefficient, A is the irradiated area of the container. T<sub>surr</sub> is the temperature of surrounding (regarded as constant). In order to get the hA, a dimensionless driving force temperature  $\theta$  is defined as follows equation 4:

$$\theta = \frac{T - T_{surr}}{T_{max} - T_{surr}} \quad (4)$$

Where T<sub>max</sub> is the equilibrium temperature and the hA can be deprived from the time constant  $\tau$  as following equation

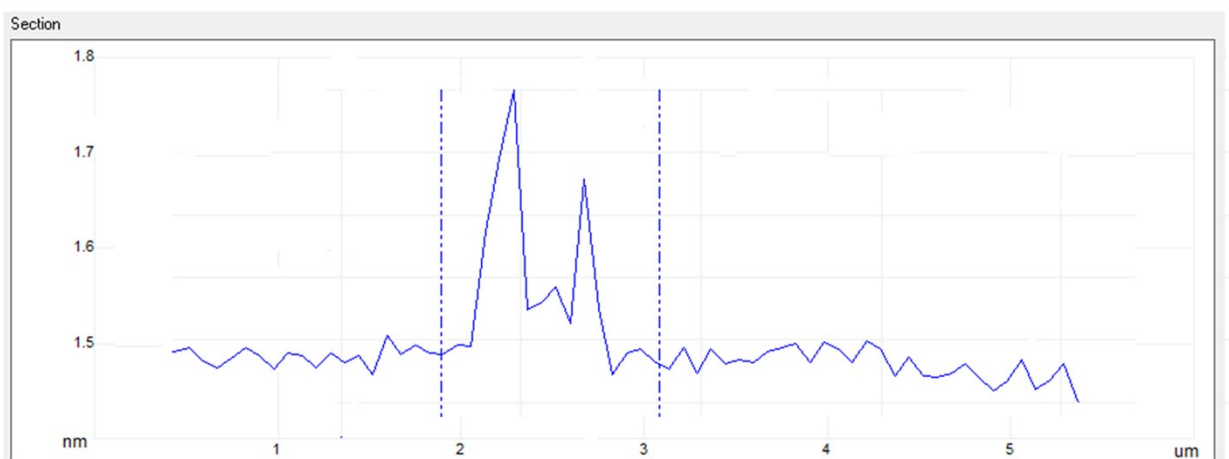
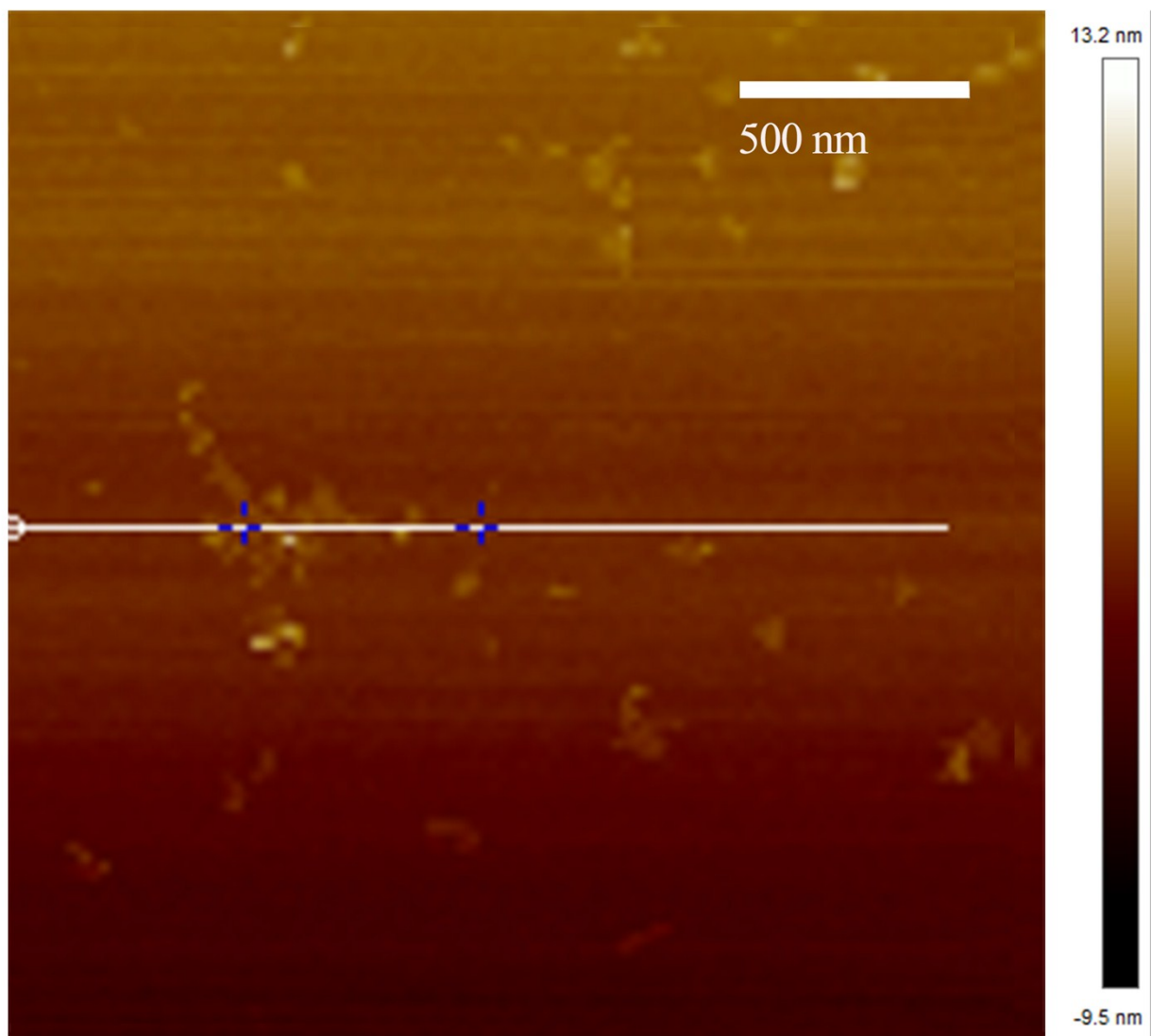
5:

$$\tau = \frac{mC_p}{hA} \quad (5)$$

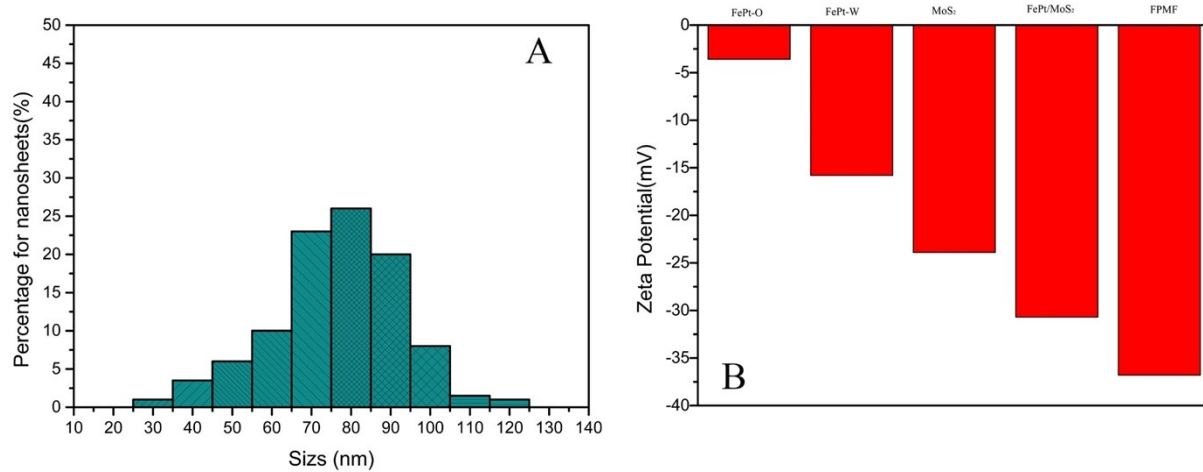
Therefore, time constant is linear-fitted to be  $\tau_s = 285.5$  s according to the linear time from the cooling period after 600 s vs  $\ln\theta$  (Figure S3). Thus, based on the equation 5, the  $hA$  is deduced to be  $16.56 \text{ mW } ^\circ\text{C}^{-1}$ . Thus photothermal conversion efficiency of FePt/MoS<sub>2</sub> NPs can be calculated to be 32.69 % by equation (2) and (1).

1. Yin W, Yan L, Yu J, Tian G, Zhou L, Zheng X, et al. ACS Nano 2014; 8: 6922-33.

2. Liu X, Li B, Fu F, Xu K, Zou R, Wang Q, et al. Dalton Transactions 2014; 43: 11709-15.



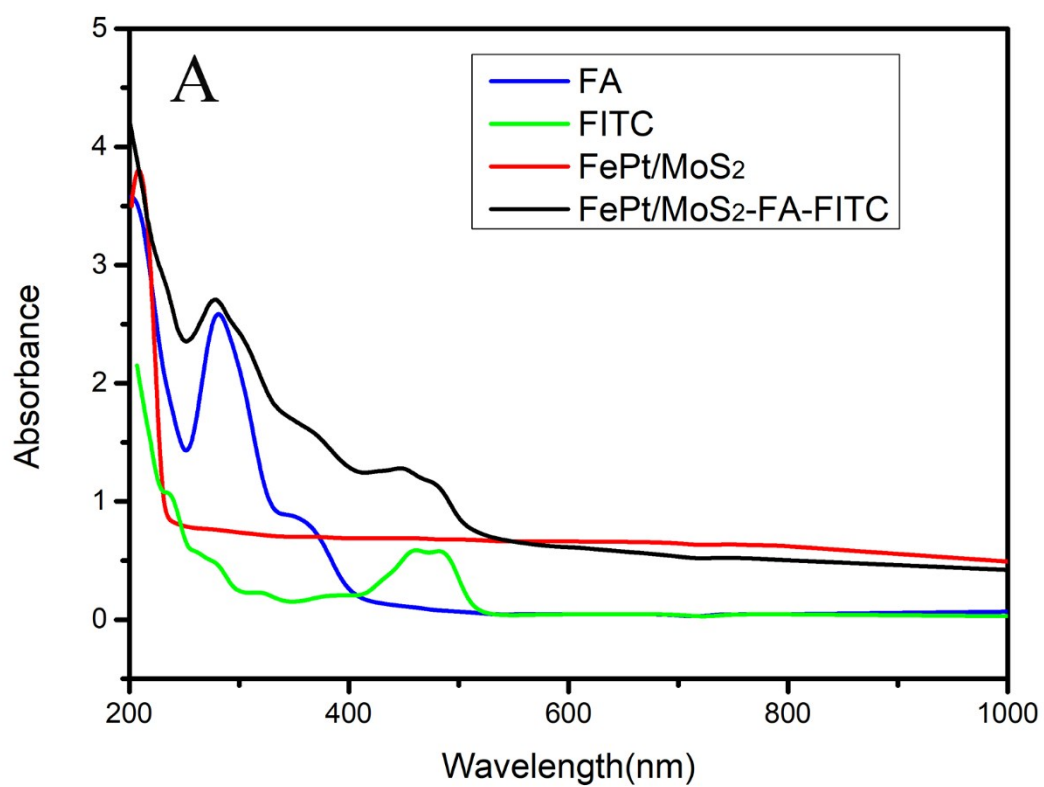
**Figure S1.** Atomic force microscopy (AFM) micrograph of MoS<sub>2</sub> nanosheets



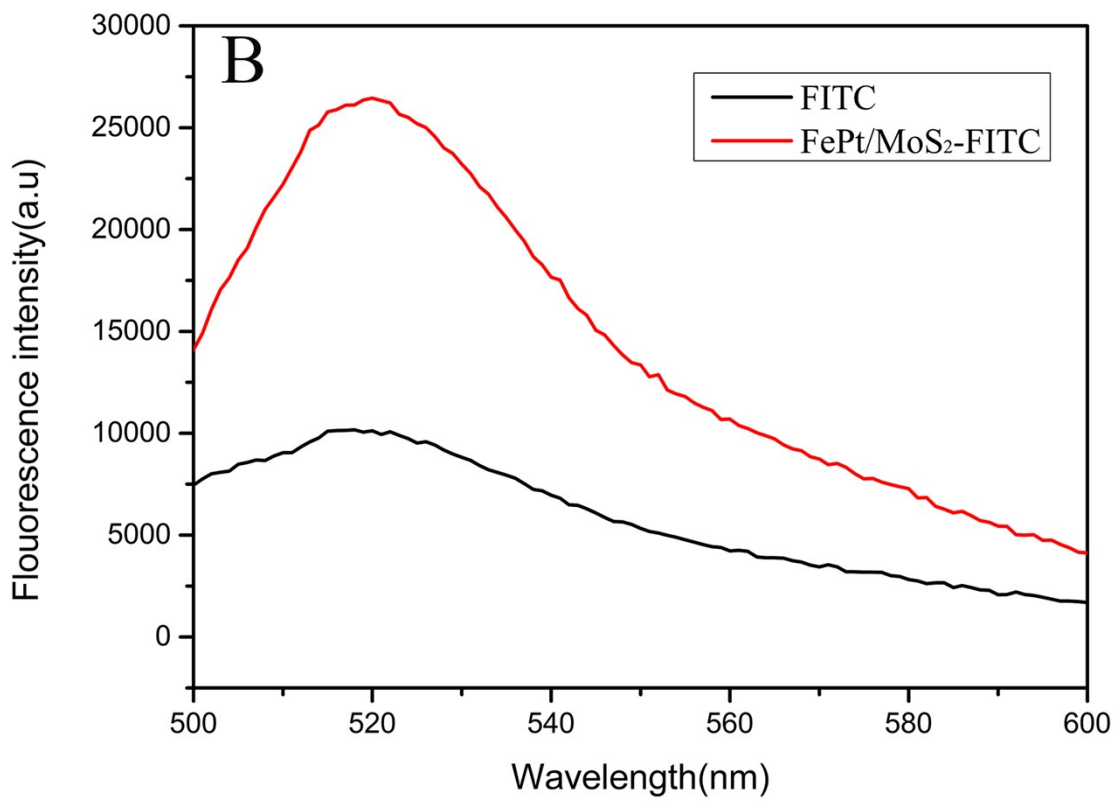
**Figure S2.** A: The size of FePt/MoS<sub>2</sub> nanocomposites. B: The Zeta of FePt-O, FePt-W, MoS<sub>2</sub>, FePt/MoS<sub>2</sub> and FePt/MoS<sub>2</sub>-FA nanocomposites solutions



**Figure S3.** Stabilities of the as-prepared FePt/MoS<sub>2</sub> were dissolved in H<sub>2</sub>O (A), PBS (B), and DMED medium (C) or FPMF NCs were dissolved in H<sub>2</sub>O (D), PBS (E), and DMED medium (F) for 72 h, respectively



**Figure S4.** UV-Vis spectra of the as-synthesized FePt/MoS<sub>2</sub>, FePt/MoS<sub>2</sub>-FA-FITC, free HS-PEG-FA and free HS-PEG-FITC



**Figure S5.** The fluorecence spectrum of FePt/MoS<sub>2</sub>-FA-FITC and free HS-PEG-FITC



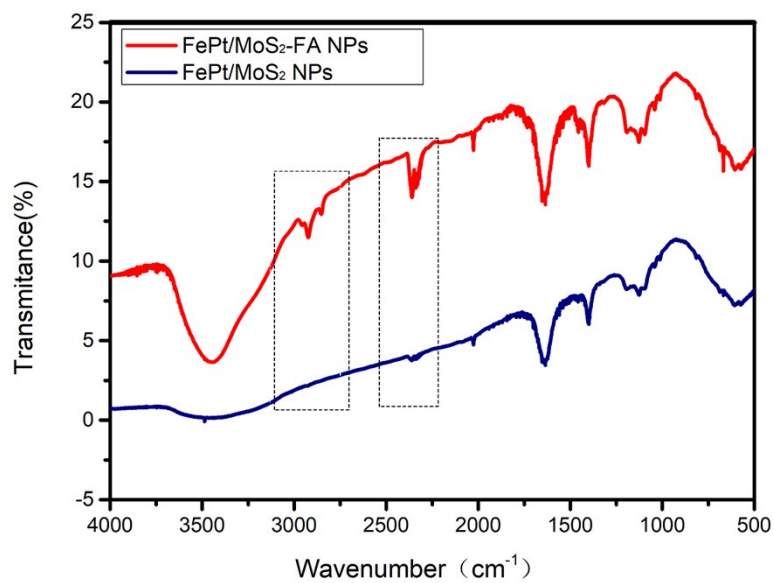


Figure S6. FT-IR of FePt/MoS<sub>2</sub> HNPs before and after conjugated with SH-PEG-FA

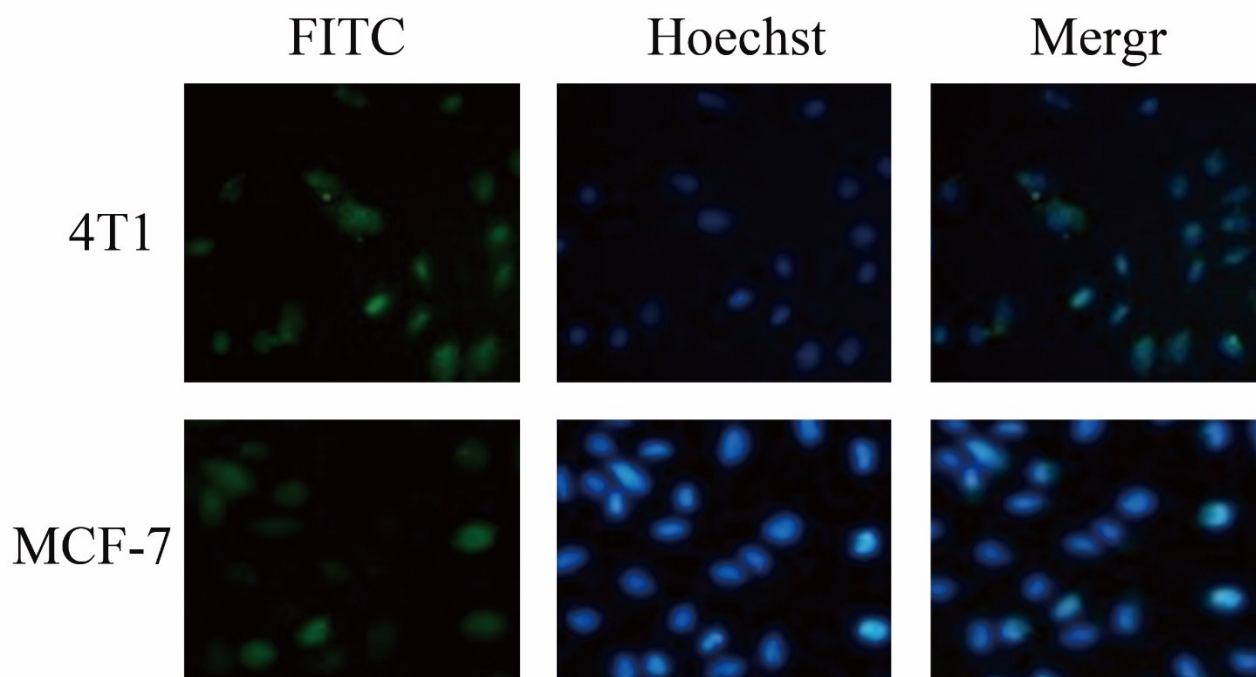
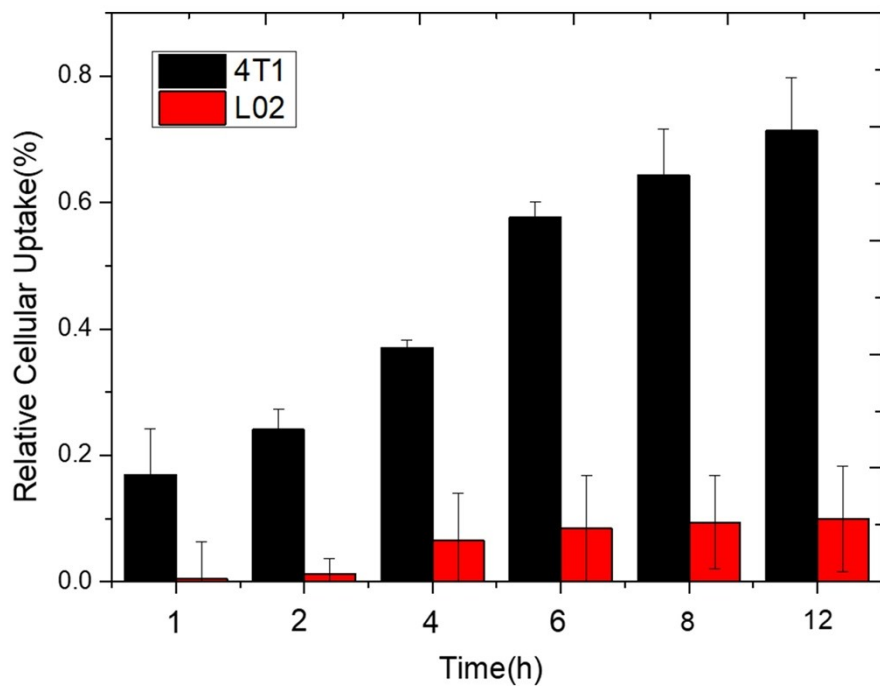
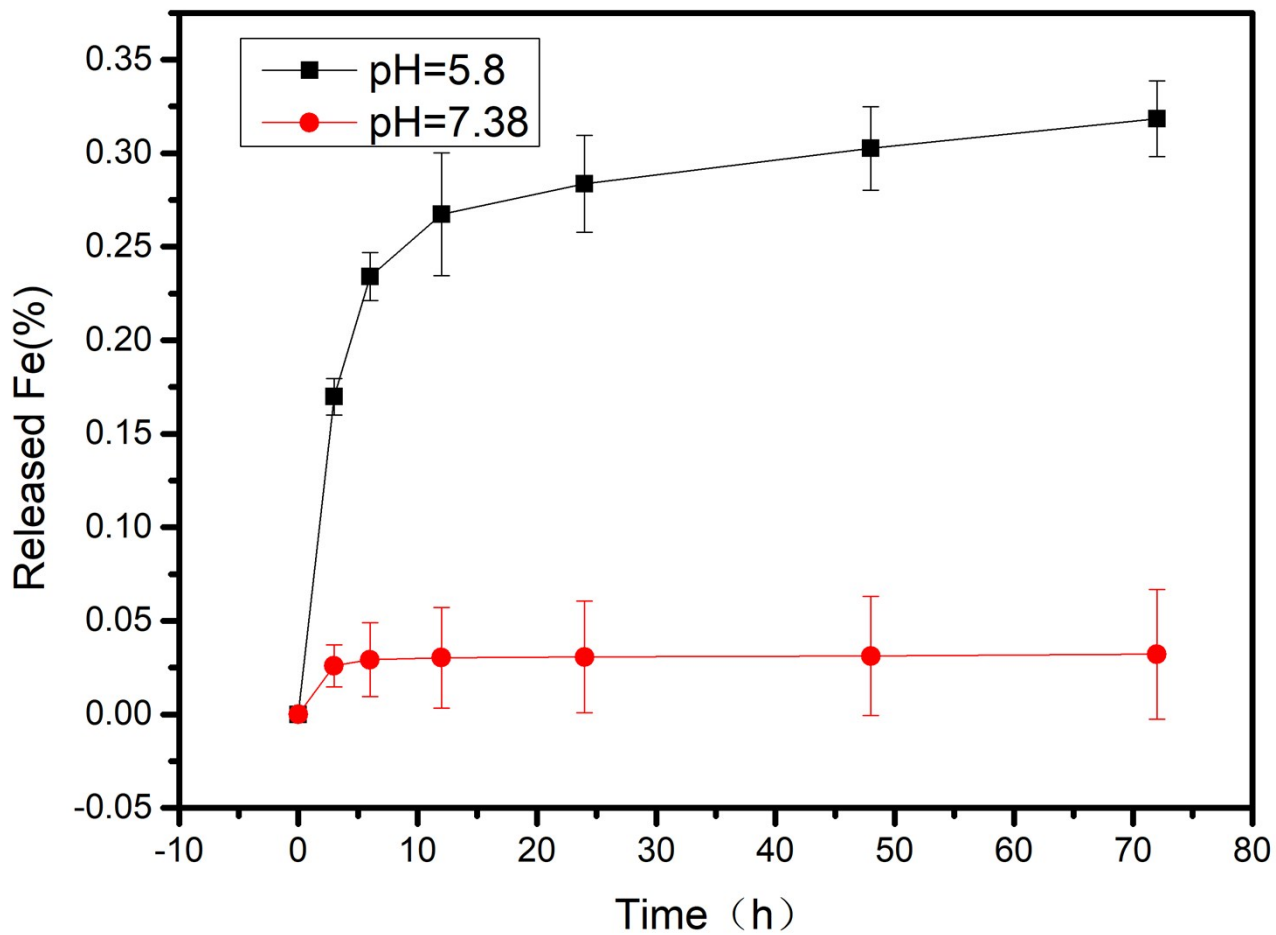


Figure S7. The fluorescence image of FePt/MoS<sub>2</sub>-FA-FITC



**Figure S8.** The relative cellular uptake of FePt/MoS<sub>2</sub> nanocomposites toward 4T1 and L02 cells under different incubation times at the Fe concentration of 80  $\mu\text{g mL}^{-1}$



**Figure S9.** Situations of Fe releasing from FePt/MoS<sub>2</sub> HNPs with time in PBS (pH=5.8 and 7.38, respectively)

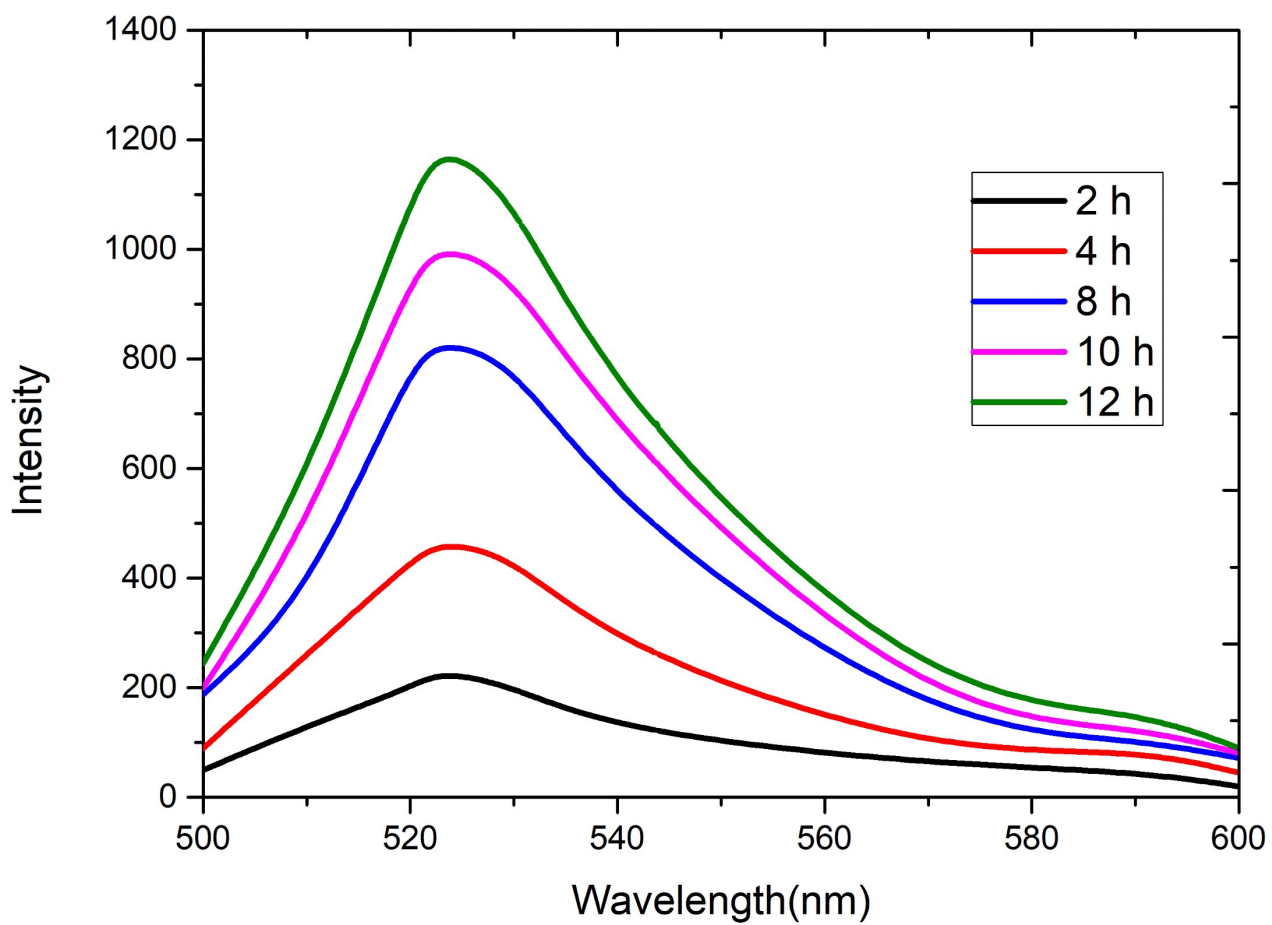
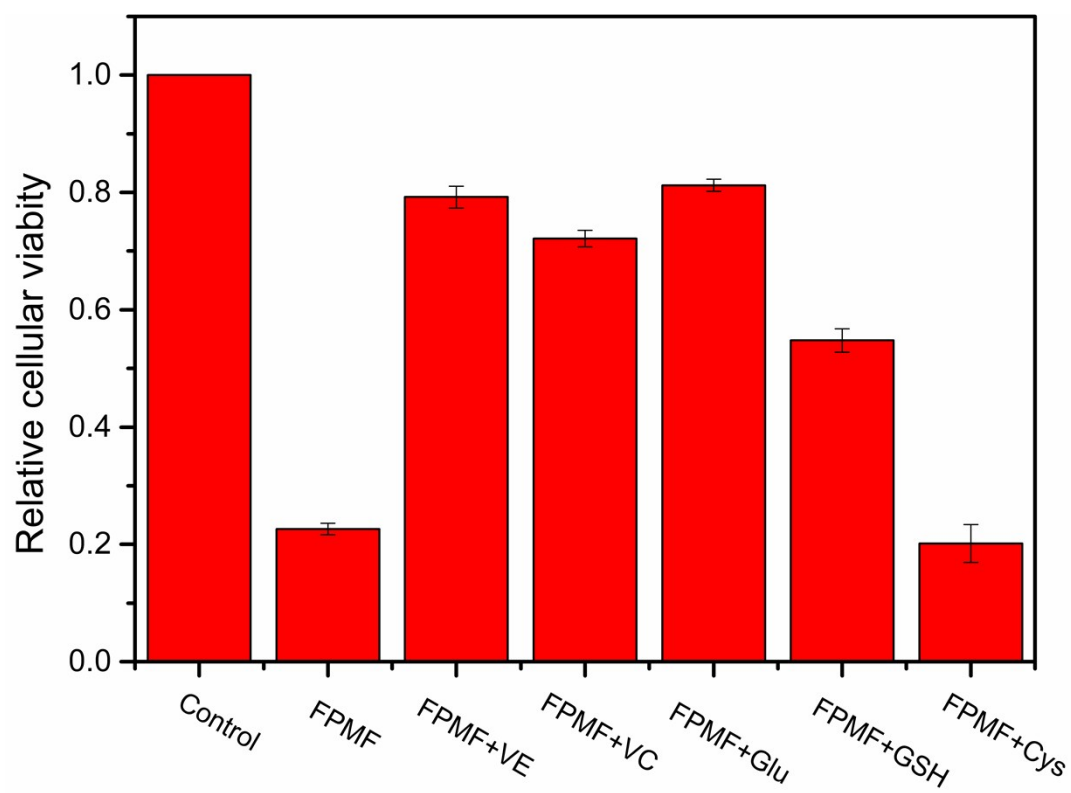
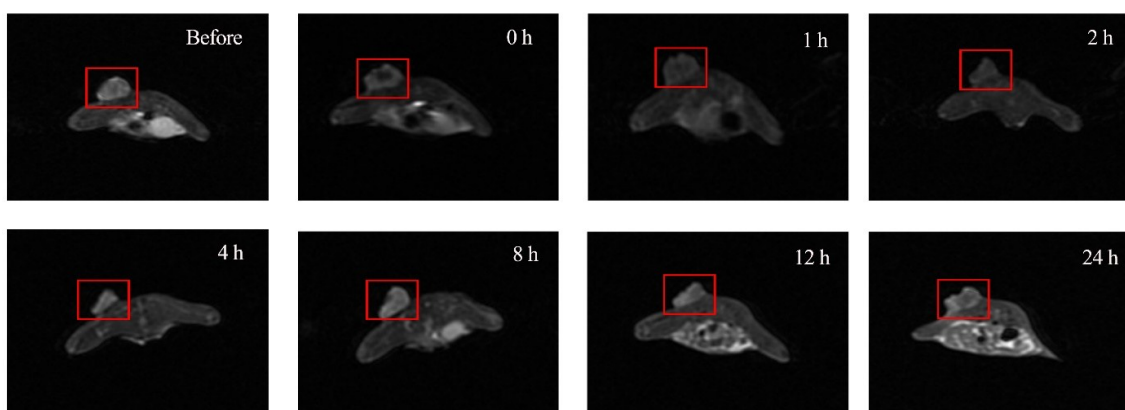


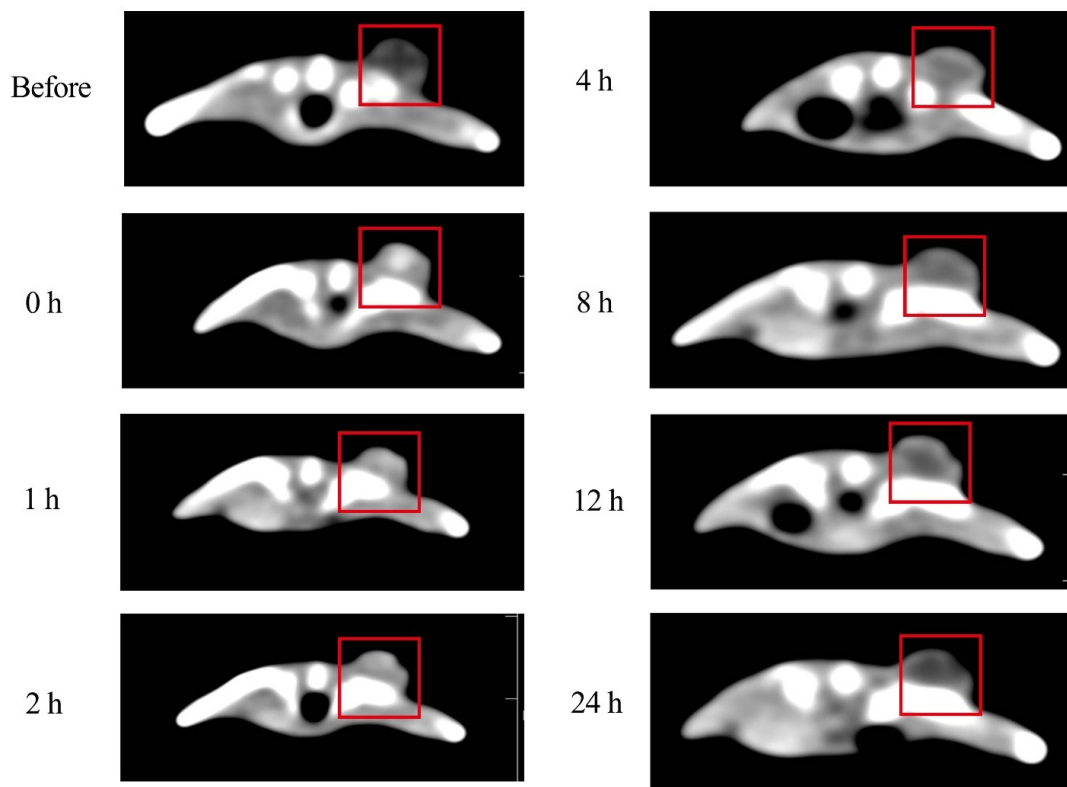
Figure S10. Time-dependent fluorescent intensity from DCFH-DA labeled 4T1 cells after being treated with FePt/MoS<sub>2</sub>-FA at the Fe concentration of 80 mg mL<sup>-1</sup>



**Figure S11.** Viabilities of MCF-7 cells processed with FPMF NCs, FPMF NCs + VE, FPMF NCs + VC, FPMF NCs + Glu, FPMF NCs + GSH and FPMF NCs + Cys

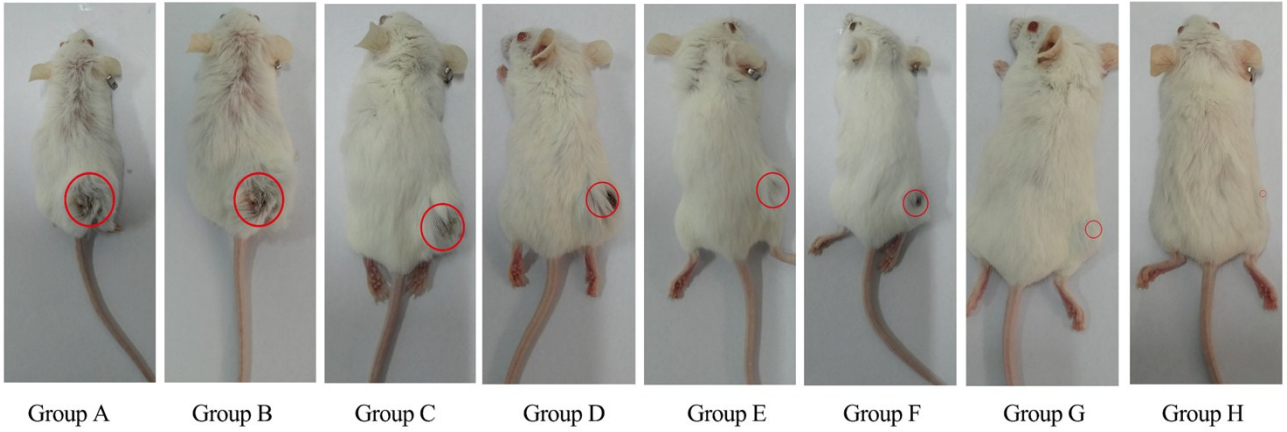


**Figure S12.** In vivo T2-weighted MR imaging (axial plane) of a 4T1 tumor-bearing mouse at different time intervals after intratumor injection with FPMF NCs. Tumor tissue was indicated with red pane

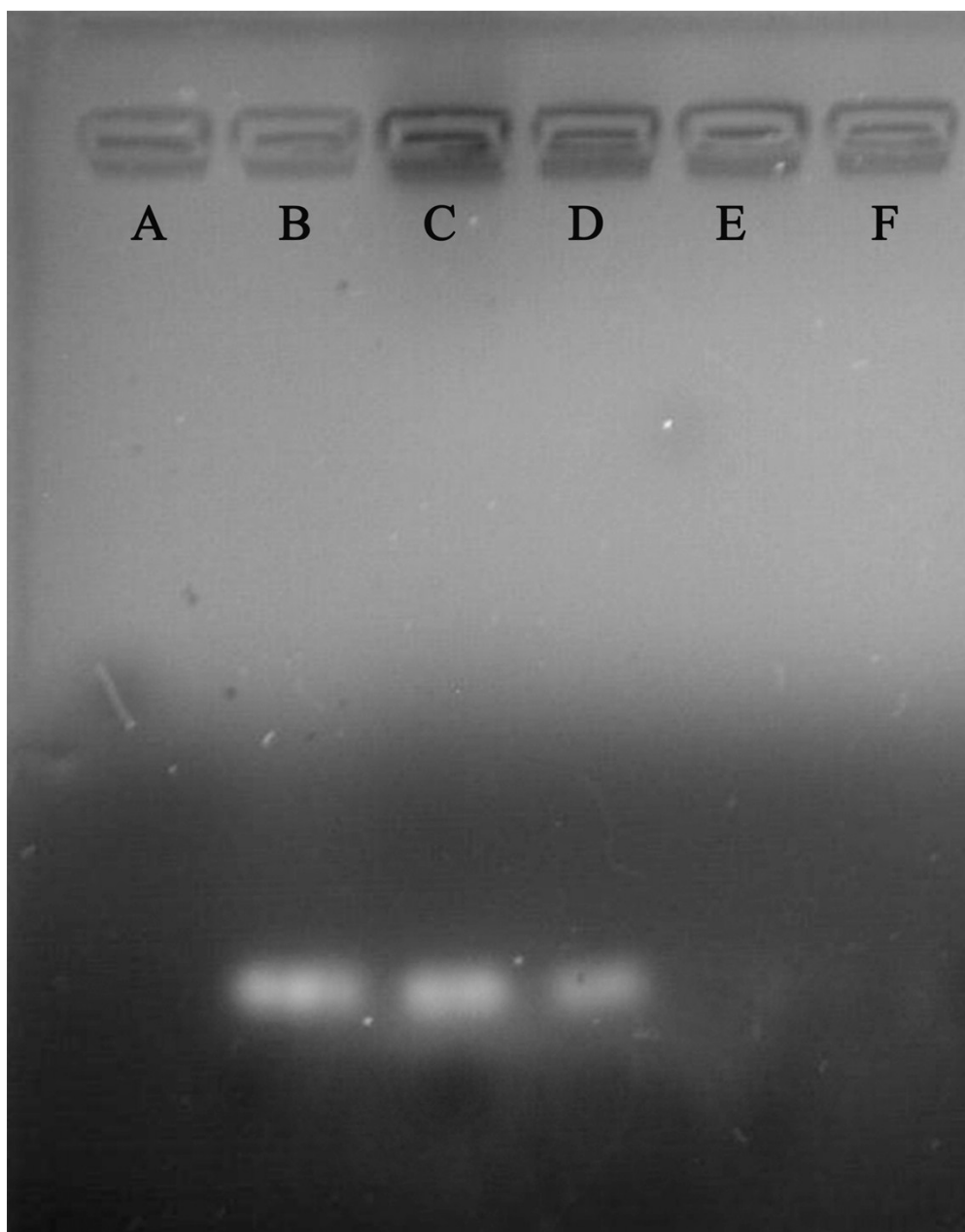


**Figure S13.** The CT images of the 4T1 tumor-bearing mouse taken before (0 h) and after (1, 2, 4, 8, 12 and 24 h) intratumor injection of FPMF NCs

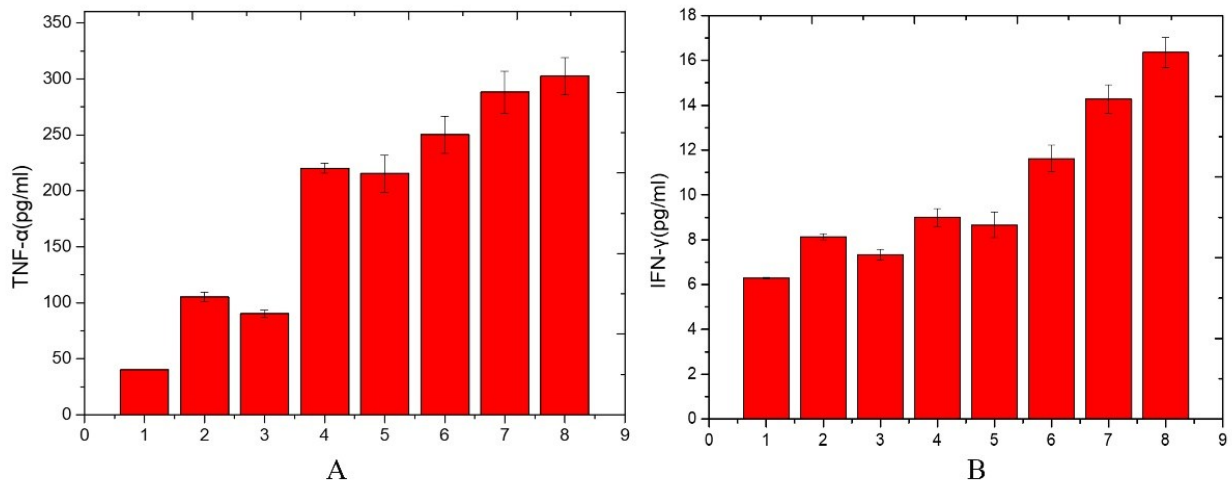




**Figure S14.** These images of mouse for different groups



**Figure S15.** Agarose gel electrophoresis images of different nanocomposites ( A: PBS; B: Free CpG ODN; C: The ratio of CpG ODN to FePt/MoS<sub>2</sub> was 1:5; D: The ratio of CpG ODN to FePt/MoS<sub>2</sub> was 1:10; E: The ratio of CpG ODN to FePt/MoS<sub>2</sub> was 1:20; F : FePt/MoS<sub>2</sub>)



**Figure S16.** Cytokine levels in sera from mice at day 22 after treatment. (Group 1: PBS; 2: PBS with CpG ODN; 3: PBS with anti-CTLA4; 4: PBS with CpG ODN with anti-CTLA4; 5: FPMF@CpG ODN NCs; 6: FPMF@CpG ODN NCs with Laser; 7: FPMF@CpG ODN NCs with anti-CTLA4; 8: FPMF@CpG ODN NCs with anti-CTLA4 with Laser. n=5)