## **Supporting Information**

## Facile synthesis of polypyrrole@metal-organic frameworks core-shell nanocomposites for dual-mode imaging and synergistic chemo-photothermal therapy of cancer cells

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Fig. S1 FTIR spectra of PPy@MIL-100(Fe) NCs and PPy NPs.



Fig. S2  $N_2$  adsorption-desorption isotherm and pore size distribution of PPy@MIL-100(Fe) NCs.



Fig. S3 XPS analysis of PPy@MIL-100(Fe) NCs.



Fig. S4 XRD patterns (a) PPy NPs and (b) PPy@MIL-100(Fe) NCs.



Fig. S5 EDX analysis of PPy@MIL-100(Fe) NCs.



**Fig. S6** (A) Photothermal effect of the irradiation of the aqueous dispersion of PPy@MIL-100(Fe) NCs with the NIR laser (808 nm, 2 W cm<sup>-2</sup>), in which the irradiation lasted for 5 min, and then the laser was turned off. (B) Time constant for heat transfer from the system is determined to be  $\tau_s = 202$  s by applying the linear time data from the cooling period versus negative natural logarithm of driving force temperature.



**Fig. S7** Photographs of PPy@MIL-100(Fe) NCs (a) water, (b) PBS buffer and (c) culture medium with serum and they were stored for 4 days, respectively.



**Fig. S8** UV-Vis spectra of PPy@MIL-100(Fe) NCs (a) and DOX-loaded PPy@MIL-100(Fe) NCs (b)



**Fig. S9** The Fe content profiles of PPy@MIL-100(Fe) NCs in the PBS (pH 5.3, pH 7.4) at different time points at 37 °C.



Fig. S10 CLSM images of HepG-2 cells incubated with DOX-loaded PPy@MIL-100(Fe) NCs for 1, 2, 8, 12 h at 37 °C, respectively. Each series can be classified to cell nucleus (being dyed in blue by DAPI), DOX-loaded PPy@MIL-100(Fe) NCs and the merged images of both above, respectively. All scale bars are 20  $\mu$ m.