## **Supporting Information**

## Polymeric <sup>1</sup>H/<sup>19</sup>F Dual-modal MRI Contrast Agent with

## **Snowman-like Janus Nanostructure**

Ziwei Duan<sup>1</sup>, Jialei Han<sup>1</sup>, Yadong Liu<sup>1</sup>, Xinyu Zhao<sup>1</sup>, Bo Wang<sup>1</sup>, Shuaishuai Cao<sup>2,\*</sup>, Dalin Wu<sup>1,3\*</sup>

Name	Reactants					Diameter	Zeta Potential
	DIMA	St	DSDA	NaVBS	KPS	(nm)	(mV)
	(mL)	(mL)	(µL)	(mg)	(mg)		
SNP-1	3.6	0.9	45	110	50	$118\pm36$	$-54 \pm 1$
SNP-2	2.25	2.25	45	110	50	$59 \pm 17$	$-45 \pm 2$
SNP-3	4.5	0	45	110	50	$123\pm39$	$-40 \pm 1$

 Table S2. JNP synthesis condition and DLS/zeta potential characterization results.

Name	Reactants		Diameter	Zeta Potential	
	SNP-1	TPMA	3-TMSPMA	(nm)	(mV)
	(g)	(mL)	(mL)		
JNP-1	0.5	0.3	0.7	$157\pm50$	-61 ± 2
JNP-2	0.5	0.4	0.6	$197\pm71$	$-22 \pm 2$
JNP-3	0.5	0.5	0.5	$331 \pm 66$	-28 ± 1







**Figure S2.** The characterization result of SNP-1, JNP-1, F-JNP and J-CA. (A) TEM image of SNP-1. (B) TEM image of JNP-1. (C) Hydrodynamic values of SNP-1, JNP-1, F-JNP and J-CA in aqueous solution. (D) Zeta potential values of SNP-1, JNP-1, F-JNP and J-CA in aqueous solution. (E) Hydrodynamic diameter values of J-CA in aqueous solution in 90 days. (F) Polydispersity values of J-CA in aqueous solution in 90 days. (G) Uv-vis spectrum of TEMPO, SNP-1, JNP-1 and J-CA in aqueous solution.



Figure S3. EDX spectrum result of J-CAs.



Figure S4. <sup>19</sup>F NMR spectrum of J-CA in  $D_2O/H_2O$  (1/9, v/v) at concentration of 5-40 mg/mL.



Figure S5. (A) <sup>19</sup>F NMR spectrum of SNP-1, JNP-1, F-JNP and J-CA in  $D_2O/H_2O$  (1/9, v/v) at concentration of 10 mg/mL. (B) T<sub>1</sub>-weighted <sup>1</sup>H MRI and <sup>19</sup>F "hot spot" MRI images of SNP-1, JNP-1, F-JNP and J-CA in  $D_2O/H_2O$  (1/9, v/v) at concentration of 20 mg/mL.



Figure S6. Fluorescence image of J-CA and J-CA-C in aqueous solution.



**Figure S7.** J-CA-C cell uptake behavior characterization. (A). Flow cytometry result at the time range of 0-6 h. (B) Mean fluorescence intensities of the 4T1 cell after incubation with J-CA-C at incubation time of 0-6 h. (C) CLSM images of uptake behavior of J-CA-C inside 4T1 cells at 2 h, 4 h, and 6 h post incubation.



**Figure S8.** Biocompatibility characterization of J-CA. (A) Digital images of hemolysis behavior of J-CA at concentration of 0.05-2 mg/mL against HUVEC. (B) Statistic result of hemolysis ratio of J-CA at concentration of 0.05-2 mg/mL against HUVEC. (C) Physiology indices values of mice on 7 days post administrating of J-CA in mice intravenously.



**Figure S9.** (A) *In vivo* fluorescence imaging and (B) quantification of average fluorescence intensity of tumor of 4T1 mice after vein injection of J-CA-Cy5.5 and free Cy5.5. (C) *Ex vivo* fluorescence images and (D) quantification of the fluorescent signals of 4T1 tumor and major organs of mice at 24 h post-injection.