## Supporting Information

## Self-driven immune checkpoint blockade and spatiotemporal-sensitive immune

## response monitoring in acute myeloid leukemia using all-in-one turn-on

## bionanoprobe

Dangui Zhang,<sup>a,b</sup> Honglian Wu,<sup>c</sup> Tianci Wang,<sup>a</sup> Yuting Wang,<sup>a</sup> Sixi Liu,<sup>a</sup> Feiqiu Wen,<sup>a\*</sup> Gerile Oudeng,<sup>a\*</sup> and Mo Yang,<sup>c\*</sup>

- <sup>a</sup> Department of Hematology and Oncology, Shenzhen Children's Hospital of Shantou University Medical College, Futian, Shenzhen, Guangdong, 518026 PR China
- <sup>b</sup> Research Center of Translational Medicine, Second Affiliated Hospital of Shantou University Medical College, Shantou, Guangdong, 515041 PR China
- <sup>c</sup> Department of Biomedical Engineering, Hong Kong Polytechnic University, Hung Hom, Kowloon,
  999077 Hong Kong SAR, PR China

\*Corresponding authors. E-mail addresses: fwen62@163.com (Feiqiu Wen), gerile.oudeng@connect.polyu.hk (Gerile Oudeng), Mo.Yang@polyu.edu.hk (Mo Yang).



Fig. S1 Size distribution of PDA, PDA–PEG, and BP-PDA by dynamic light scattering (DLS)



Fig. S2 X-ray photoelectron spectra of PDA (A), PDA-PEG (A & B), and BP-PDA (A & C)



Fig. S3 Fourier transform infrared spectra of PDA, PDA–PEG, and BP-PDA



**Fig. S4** The hydrodynamic diameter of PDA–PEG and B-PDA after incubation with different concentrations of bovine serum albumin (A) or serum (B) in PBS at 37°C for 3 h



**Fig. S5** Relative fluorescence intensity of BP-PDA bionanoprobes against GrB, GrA, MMP-2, and GSH. Data represented as mean  $\pm$  standard deviation (n = 3) and analyzed by t-test for two groups

comparison and one-way ANOVA for three groups comparison.



Fig. S6 Ex vivo fluorescence images of tumors and organs (kidneys, lungs, spleens, livers, and hearts)

for humanized AML mice injected with fluorescein 5-isothiocyanate (FITC) -labeled B-PDA for 24 and 48 h  $\,$ 



Fig. S7 Hematoxylin and eosin staining of main organs of mice after different treatments (Scale bars: 100  $\mu$ m)



**Fig. S8** Cellular uptake of Fluorescein 5-isothiocyanate (FITC)-labeled B-PDA (FITC@B-PDA) by THP-1, HL-60, and Ea.hy926 cells after 12 h of coculture by flow cytometry



**Fig. S9** Cellular uptake of FITC@B-PDA by THP-1 cells after coculture at different time points (4 h, 8 h, 12 h, and 24 h; scale bar:  $10 \mu m$ ) (A) and fluorescence analysis (B)



**Fig. S10** Granzyme B secretion after B-PDA treatment. Jurkat T cells were pre-treated by PMA (Phorbol 12-myristate 13-acetate) for 24 h and then cocultured with THP-1 cells for 24 h. Level of Granzyme B was measured by ELISA in supernatant.



**Fig. S11** Percentage of human CD45+CD3+CD4+/CD8+ T cells harvested from peripheral blood at 14 days after transplantation of PBMC as determined by flow cytometry



**Fig. S12** Confirmation of acute myeloid leukemia mice model. (A) Expression detection of CD11b and CD14 in THP-1 cells by flow cytometry; (B)Percentage of human CD11b+CD14+ AML cells harvested from tumor tissue in humanized AML model as determined by flow cytometry



Fig. S13 Images of mice tumor in each group after treatment



Fig. S14 MFI analysis of CD3/CD8 in tumor sections from mice with their corresponding treatments

Complex	Added	Detected	Recovery	Relative Standard
Sample	Concentration	Concentration	Rate (%)	Deviations RSD (%)
	(nM)	(nM)		
1	0	0.03	88.02	1.76
	0.3	0.32	85.36	2.25
	1	0.94	102.50	2.65
	5	4.87	94.02	1.62

Table. S1 Concentration recovery analysis of GrB target