Supporting Information

Magnetical hollow micro-sized nanoaggregates for synergistically accelerating PET glycolysis

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Total number of Tables:1 (Table S1)

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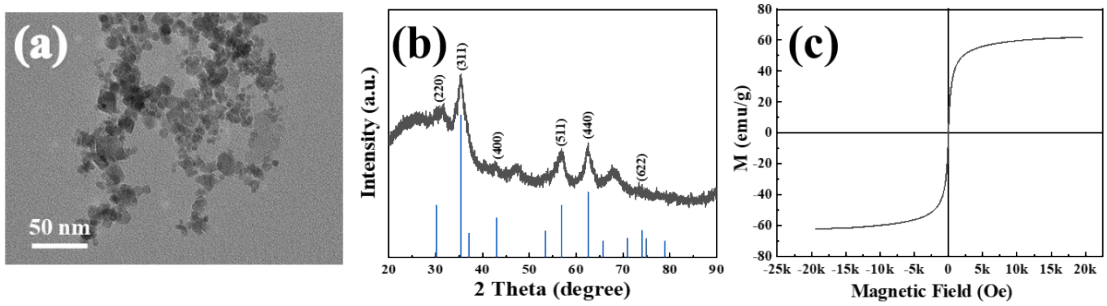

Fig. S1. (a) The TEM image, (b) XRD pattern and (c) hysteresis loop of Fe_3O_4 NPs.

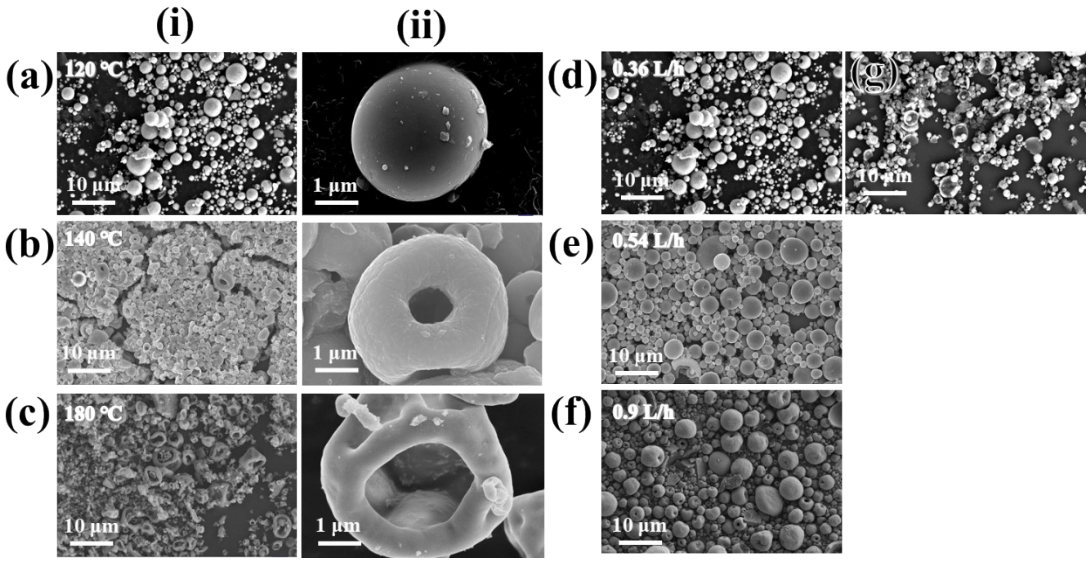

Fig. S2. SEM images of ZnO-3.5 nm-Fe_3O_4 HMNAs obtained under different inlet temperatures: (a) 120 ℃, (b) 140 ℃, (c) 180 ℃ and different feed rates: (d) 0.36 L/h, (e) 0.54 L/h, (f) 0.9 L/h. (g) The SEM image of cracked hollow microspheres after the ultrasound treatment for a period of time.

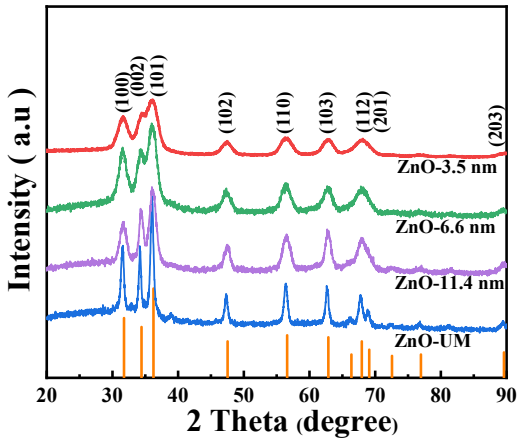

Fig. S3. XRD patterns of obtained ZnO NPs.

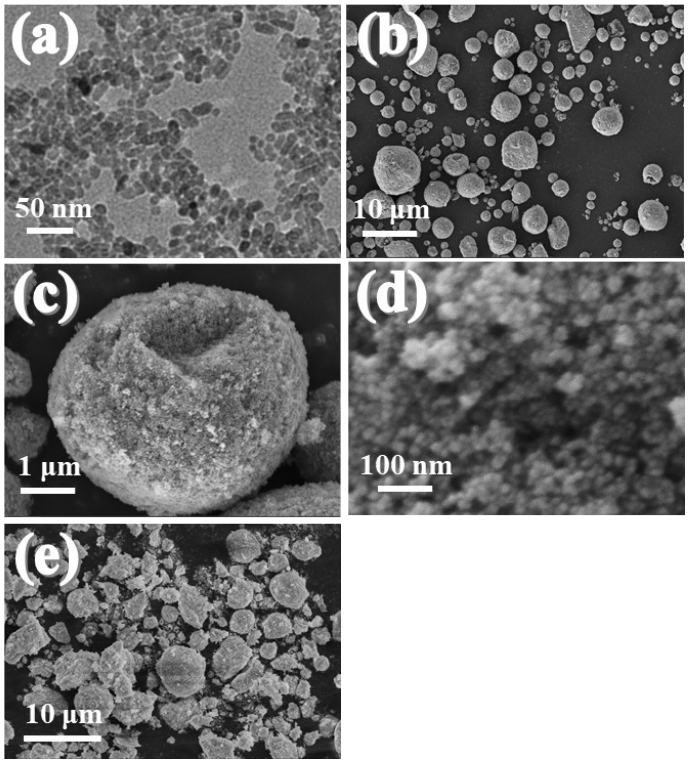

Fig. S4. (a) TEM images of the ZnO-UM suspension. (b-d) SEM images of the ZnO-UM-Fe_3O_4-2:1 HMNAs. (e) The SEM image of ZnO-Fe_3O_4 particles obtained from poorly dispersed ZnO powder.

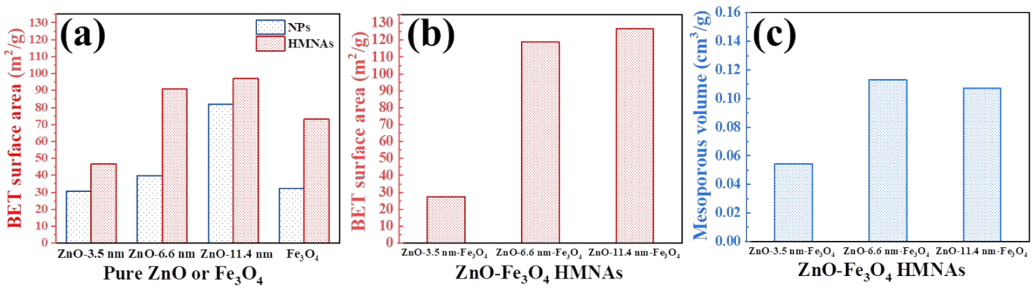

Fig. S5. BET surface areas of (a) ZnO NPs and HMNAs, Fe_3O_4 NPs and HMNAs, (b) ZnO-Fe_3O_4 HMNAs obtained from different primary sizes of ZnO. (c) Mesoporous volumes of ZnO-Fe_3O_4 HMNAs.

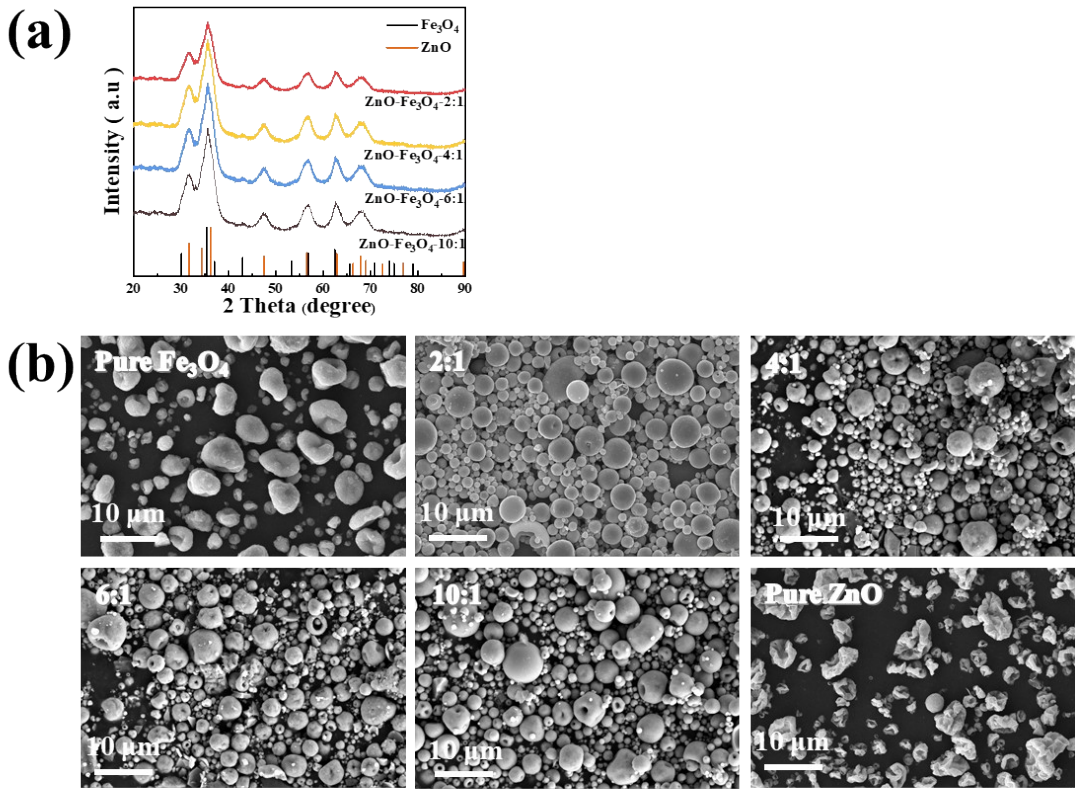

Fig. S6. (a) XRD patterns of ZnO-Fe_3O_4 HMNAs with different weight ratios of ZnO and Fe_3O_4 NPs

and (b) SEM images.

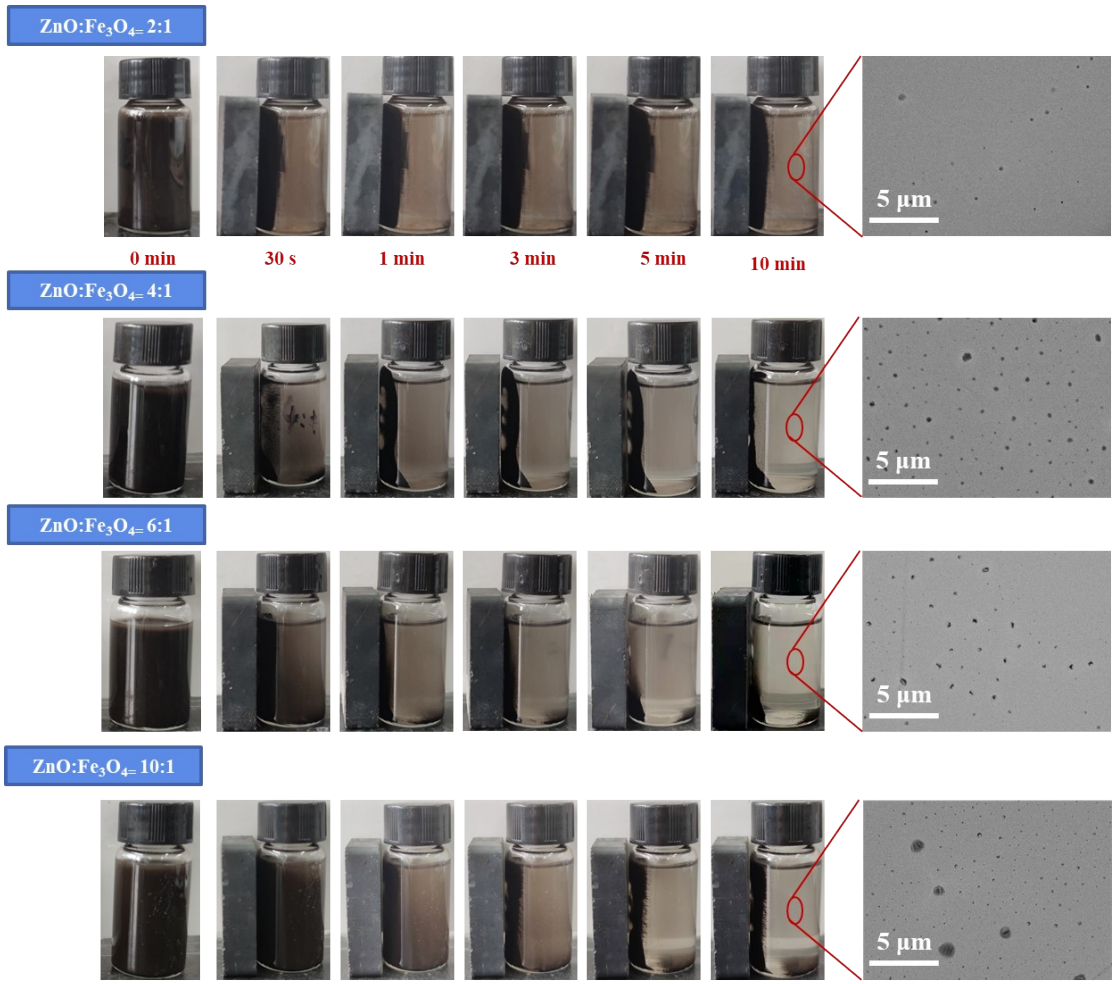

Fig. S7. Digital photographs of ZnO-Fe_3O_4 HMNAs with different weight ratios of ZnO and Fe_3O_4 NPs attracted to a magnet (3000 Gs).

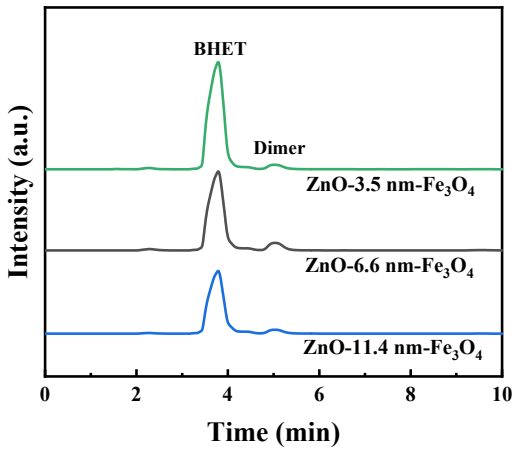

Fig. S8. HPLC chromatograms of the glycolysis products catalyzed by (a) ZnO-3.5 nm-Fe_3O_4 HMNAs, (b) ZnO-6.6 nm-Fe_3O_4 HMNAs, (c) ZnO-11.4 nm-Fe_3O_4 HMNAs.

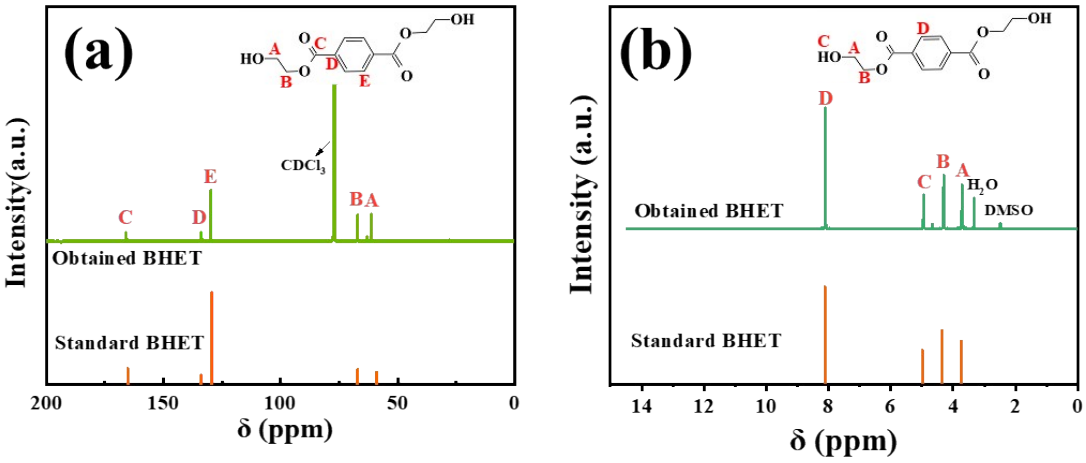

Fig. S9. (a)^{13}C NMR and (b) ^{1}H NMR spectra of obtained BHET and standard BHET.

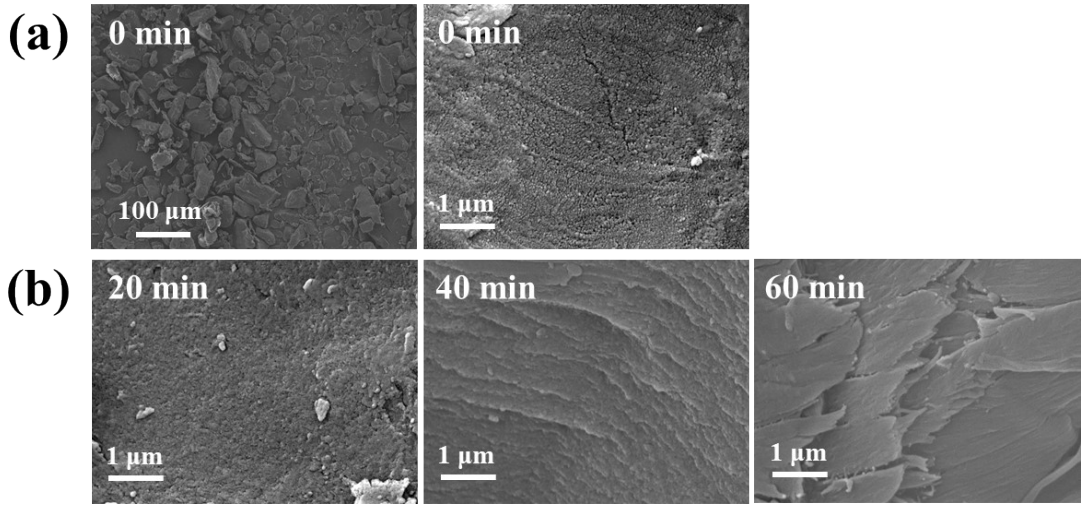

Fig. S10. SEM images of (a) virgin PET, (b) residual PET after the glycolysis for different times at 170 ℃.

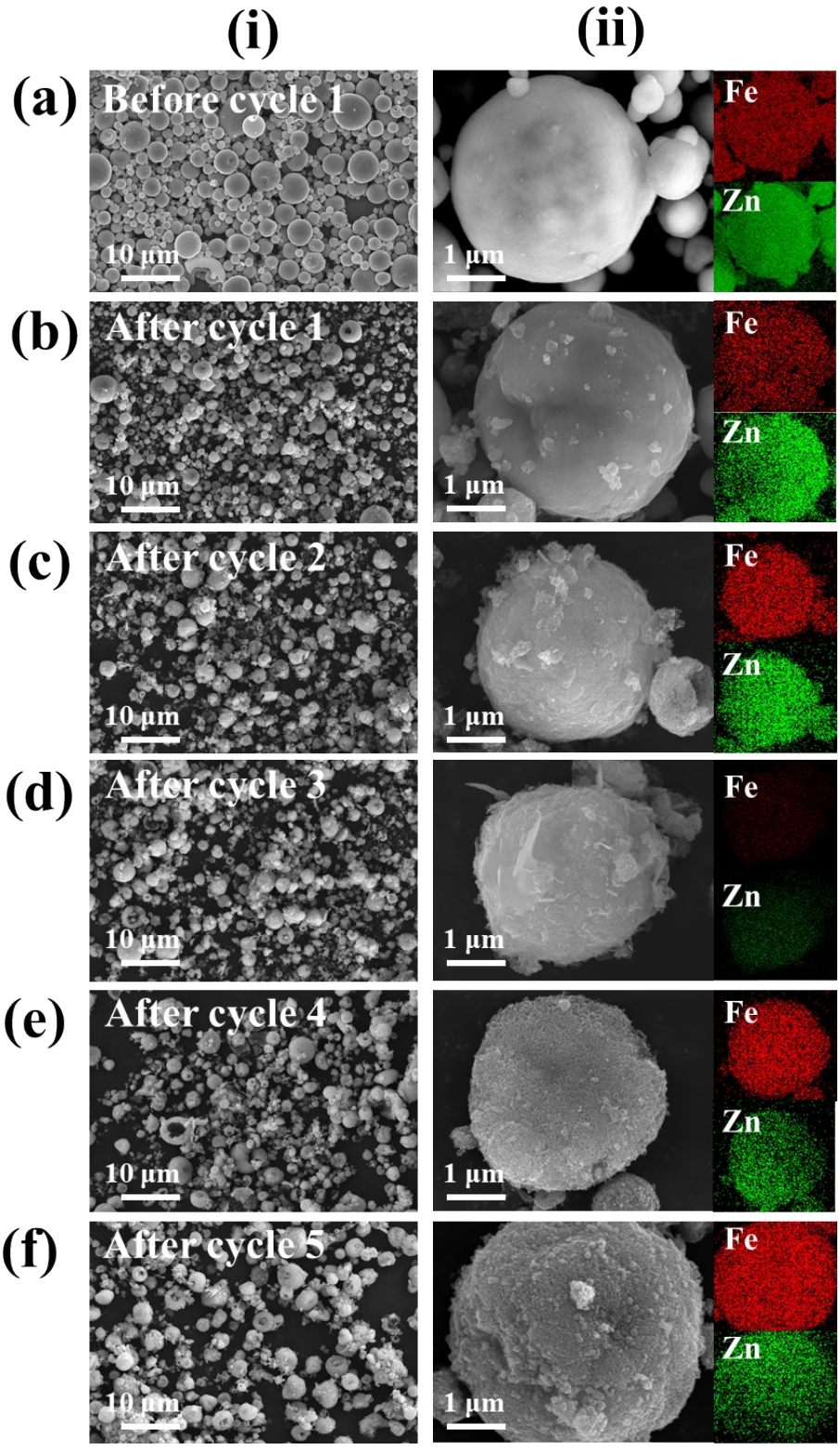

Fig. S11. SEM images, elemental mapping and EDS contents of Zn and Fe over the ZnO-Fe_3O_4 HMNAs before (a) and after each recycling step (b-f).

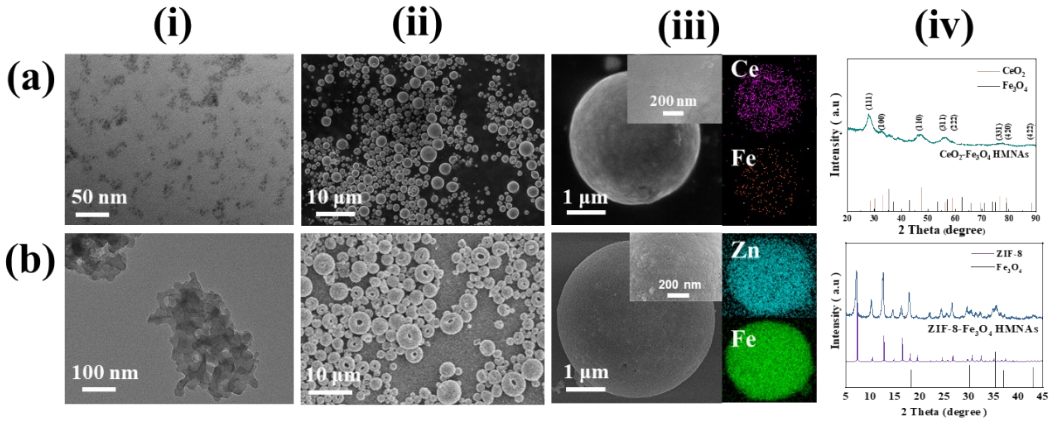

Fig. S12. (a) CeO_2-2.7 nm and CeO_2-2.7 nm-Fe_3O_4: (i) TEM image of CeO_2 nanoparticles. (ii–iii) SEM images of CeO_2-2.7 nm-Fe_3O_4 and elemental mapping**.** (iv) XRD patterns. (b) ZIF-8-30 nm and ZIF-8-Fe_3O_4: (i) TEM image of ZIF-8-30 nm. (ii–iii) SEM images of ZIF-8-30 nm-Fe_3O_4 and elemental mapping. (iv) XRD patterns.

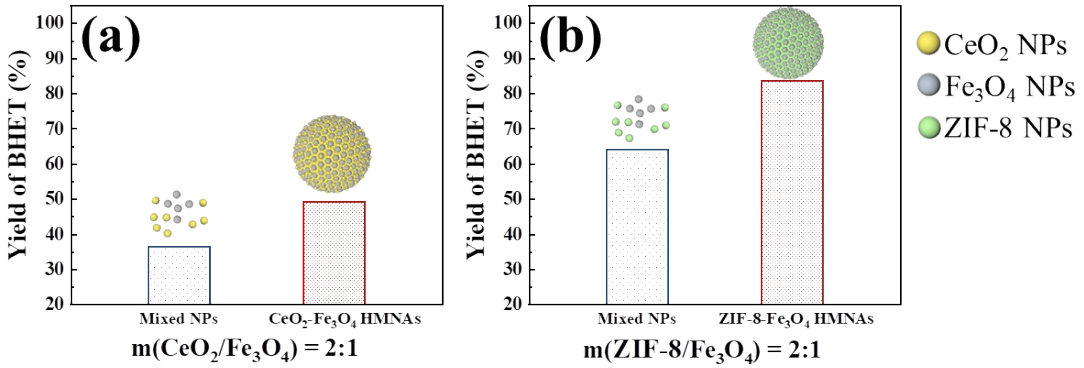

Fig. S13. (a) CeO_2-2.7 nm and Fe_3O_4 NPs directly mixed with the ratio of 2:1 and CeO_2-Fe_3O_4 HMNAs, (b) ZIF-8-30 nm and Fe_3O_4 NPs directly mixed with the ratio of 2:1 and ZIF-8-Fe_3O_4 HMNAs. (glycolysis at 180 °C, 40 min, the weight ratio of EG to PET of 6, catalysts of 1 wt%)

Table S1. Comparison of reported catalysts for the glycolysis of PET.

	Catalysts	Mass ratio of cat./PET (%)	Temperature (°C)	Time (h)	Product yield (%)	WHSV ($g_{BHET}·g_{cat}^{-1}·h^{-1}$)	Ref.
Nonmagnetic catalysts	ZnO/SBA-15	5	197	1	91	7.16	22
	rGO\[TESPMI]$_2$CoCl$_4$	0.15	190	3	95.2	83.21	46
	Graphitic carbon nitride colloid	2.5	196	0.5	80.3	25.27	12
	MAF-6	1	180	4	81.7	8.03	45
	Ultrasmall Co NPs	1.5	180	3	96	8.39	11
	ZnO nanodispersion	0.7	170	1	82.3	46.24	13
Magnetic catalysts	**ZnO-Fe_3O_4 HMNAs**	1	190	0.5	92.3	72.61	This work
	CeO_2-Fe_3O_4 HMNAs	1	197	0.75	95.39	50.03	This work
	ZIF-8-Fe_3O_4 HMNAs	1	190	0.33	85.2	101.55	This work
	$CoFe_2O_4$@ZIF-8/ZIF-67	1	200	1	84.3	33.16	16
	$CoFe_2O_4$/C10-OAC	2	195	2.5	95.4	7.50	43
	γ-Fe_2O_3/nitrogen-doped graphene	10	195	3	100	1.31	14
	Mg-Al-O@Fe_3O_4	0.5	240	1.5	80	41.96	17
	Zn-MNPs	0.8	196	2	79.8	19.62	44
	Fe_3O_4 NPs@h-BNNS	0.2	200	5	100	39.33	18