Supplementary Information

Dioxygen-Triggered Oxidation of Benzylic C-H Bonds: Insight of the Synergistic Effect of Cu-Fe Bimetallic Oxide

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**Table S1.** Comparison of lattice parameters of Fe_2O_3 in different catalysts.

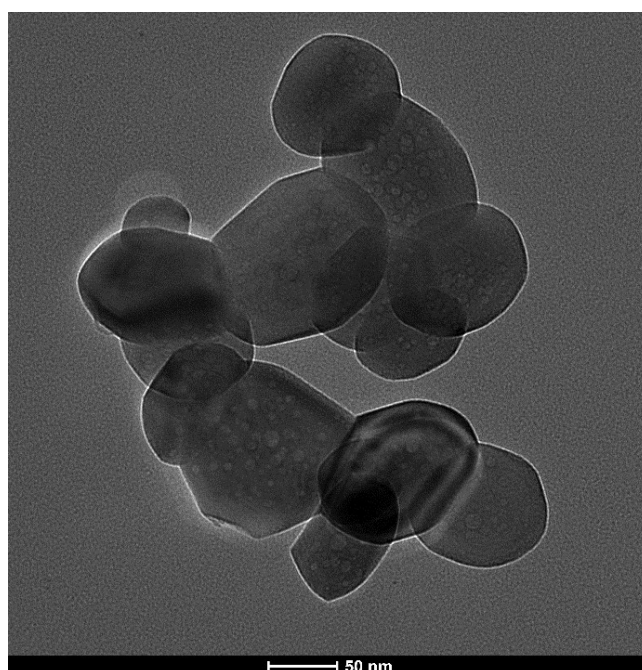| Sl. No.	Material	a=b (Å)	c (Å)	JCPDS a (Å)	JCPDS c (Å)	Error (%) a	Error (%) c
1	α-Fe_2O_3	5.02119	13.73262	5.028	13.73	0.13544	-0.0191
2	Cu1Fe2	5.02462	13.73622	5.028	13.73	0.06722	-0.0453
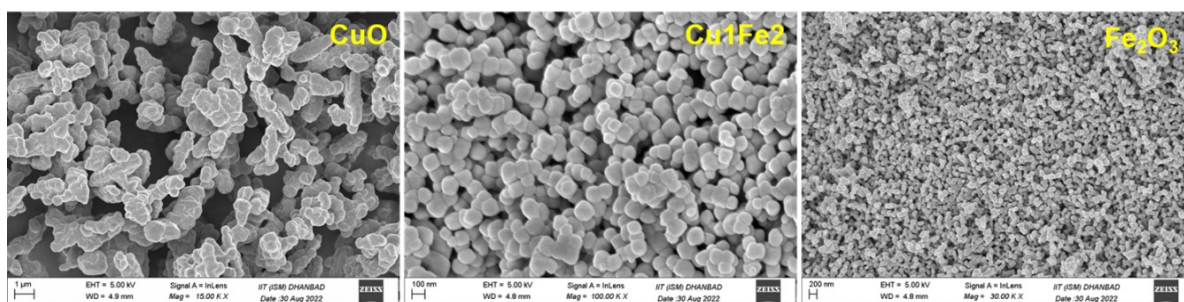
Fig. S1 FESEM images of as-synthesized catalysts.
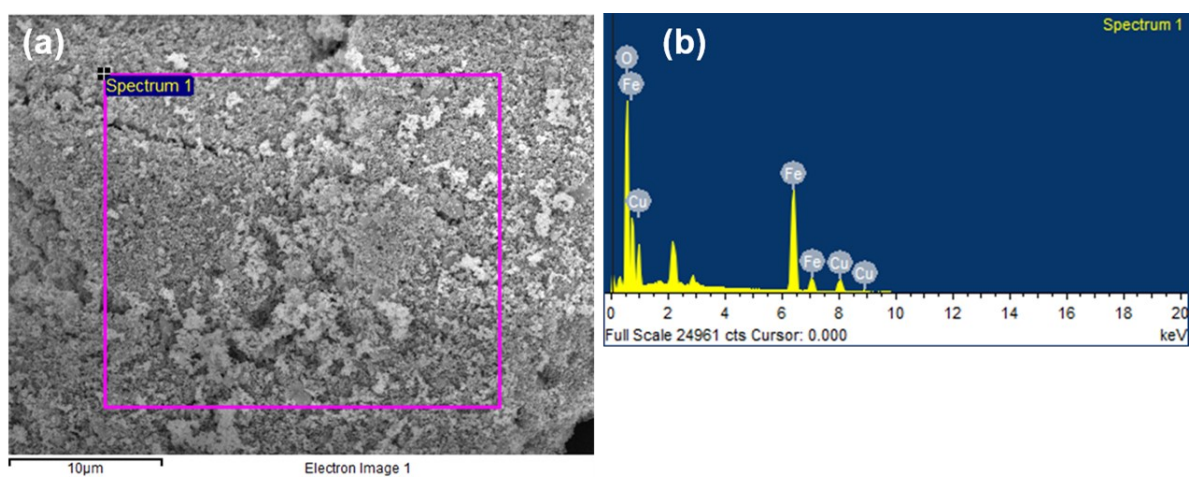
Fig. S2 Energy Dispersive X-ray Analysis (EDAX) for the elemental analysis of the Cu1Fe2 catalyst.
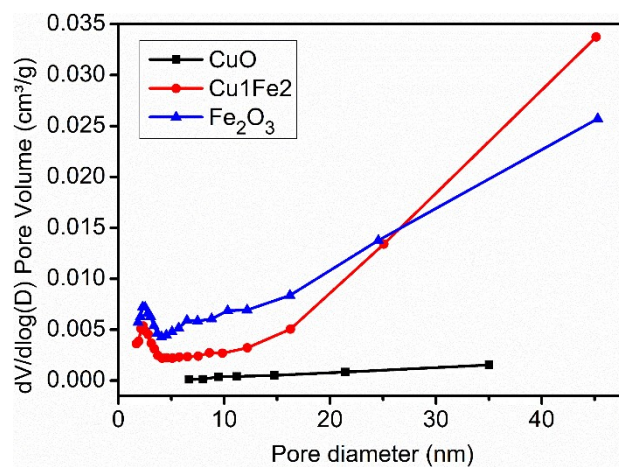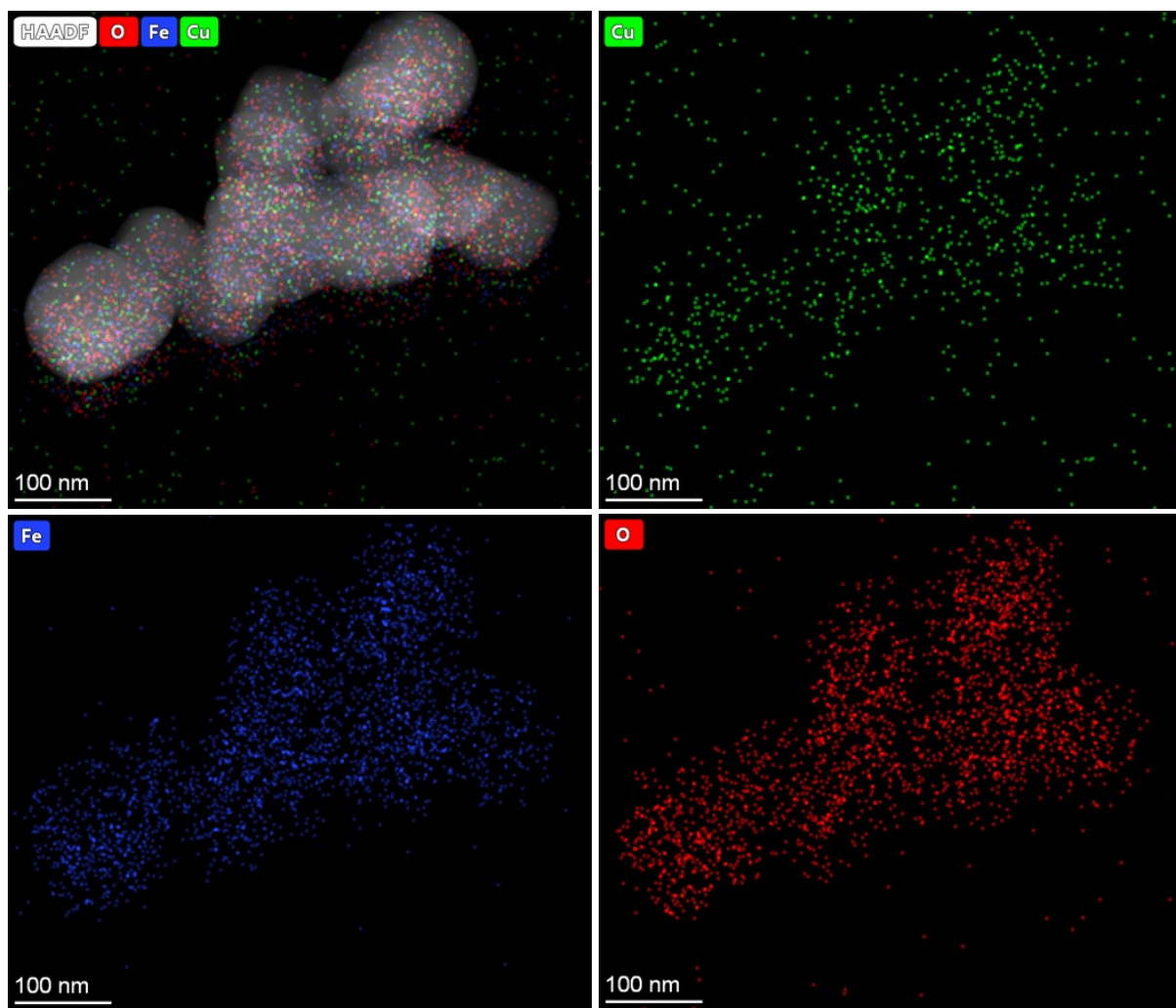
Fig. S3 BJH pore size distribution of the catalysts.

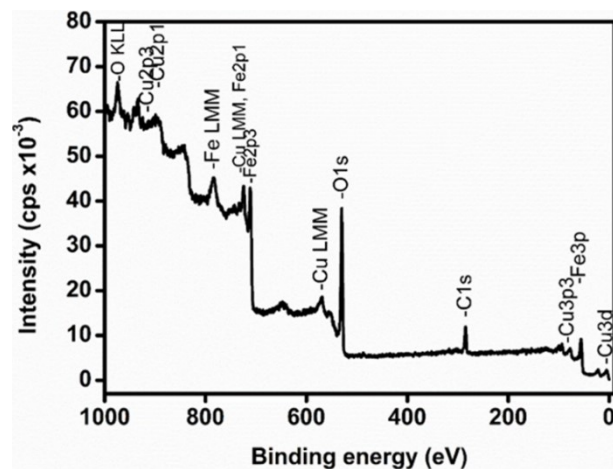

Fig. S4 TEM image of Fe$_2$O$_3$.
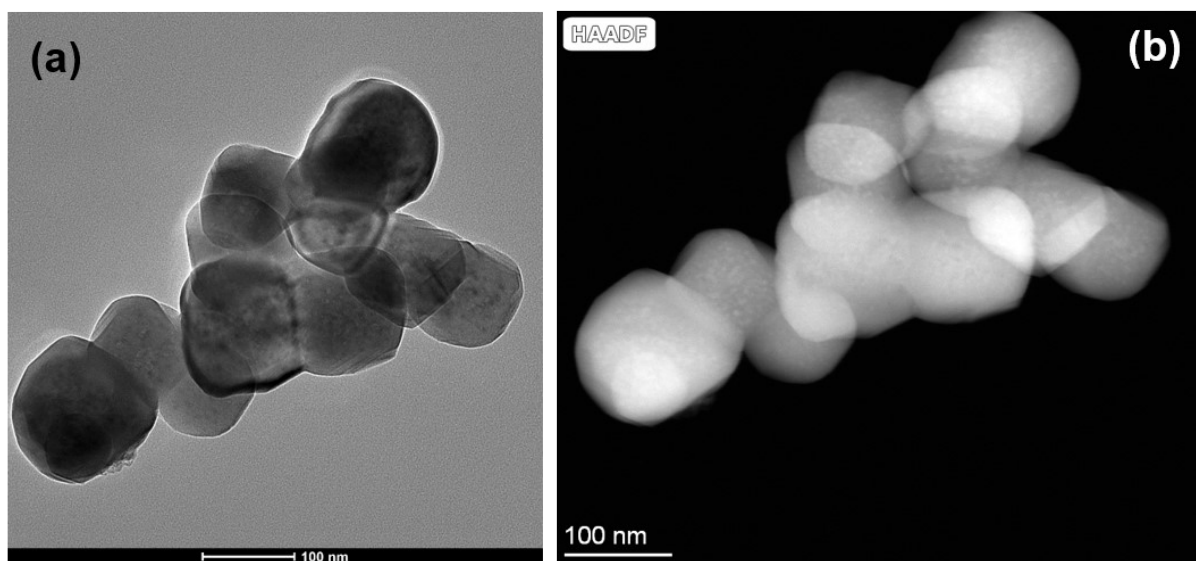
Fig. S5 (a) TEM image, (b) high-angle annular dark field (HAADF) image of as-synthesized Cu1Fe2 catalyst.

Fig. **S6** TEM-EDS mapping images of as-synthesized Cu1Fe2 catalys

Fig. S7 XPS spectra of (a) Cu 2p and (b) O 1s of CuO.

Table S2. The relative atomic concentration (%) of Cu, Fe and O and their binding energies estimated from XPS data.

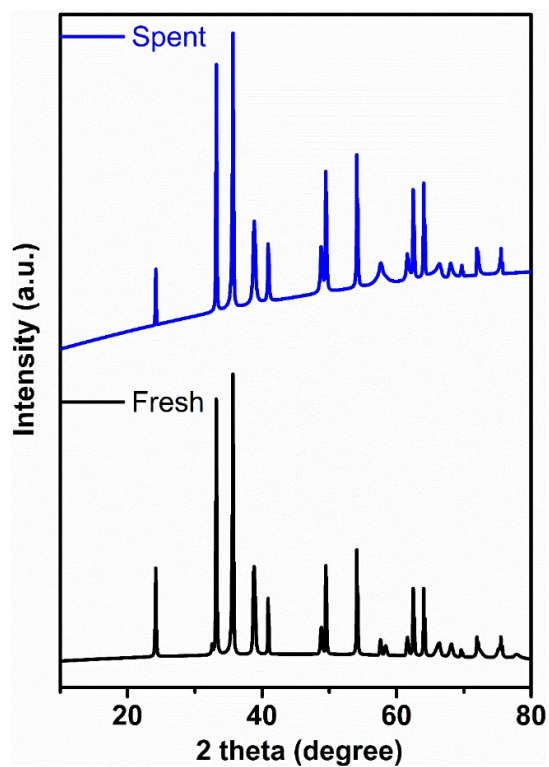| Catalyst	iron (%)				copper (%)					oxygen (%)		
	Fe^{2+}	Fe^{3+}_{octa} (2p3/2)	Fe^{3+}_{tetra} (2p3/2)	satellite	Cu^+	Cu^{2+}_{octa} (2p3/2)	$Cu^{2+}_{tetra\ (2p3/2)}$	satellite1	satellite2	O_L	O_V	O_{ads}
Fe_2O_3	-	10.5 (710.2)	7.8 (712.4)	3.2 (719.0)	-	-	-	-	-	6.1 (529.2)	19.8 (531.1)	9.1 (532.7)
Cu1Fe2-fresh	0.5 (708.3)	16.8 (710.3)	6.3 (712.6)	4.8 (718.5)	0.8 (931.7)	2.6 (933.4)	1.0 (935.7)	1.2 (940.3)	1.1 (943.2)	14.7 (528.5)	34.8 (530.2)	6.5 (532.1)
Cu1Fe2-spent	1.9 (707.4)	11.2 (710.4)	4.2 (712.8)	4.6 (719.0)	0.6 (931.6)	4.0 (932.9)	0.4 (936.2)	1.3 (939.4)	1.3 (942.3)	12.6 (528.3)	29.2 (530.0)	5.8 (532.0)
CuO	-	-	-	-	0.7 (932.4)	6.8 (933.5)	1.7 (935.7)	3.1 (941.3)	1.3 (943.8)	9.8 (529.4)	12.7 (531.5)	4.1 (533.4)
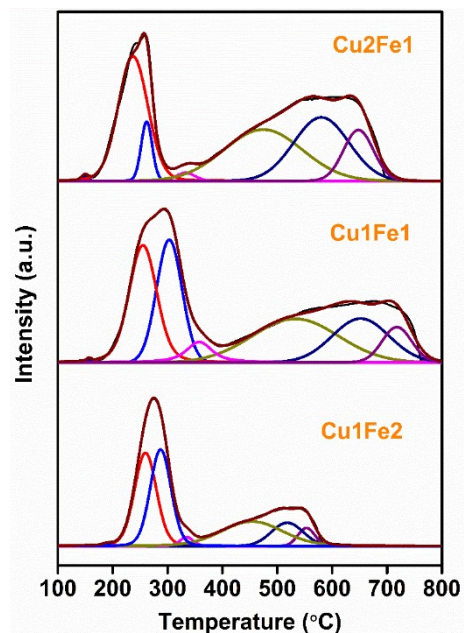
**Fig. S8** H$_2$-TPR profiles of the as-synthesized catalyst

Table S3. The deconvoluted TPR results of the catalysts.

Catalyst	Quantity (cm^3/g STP)*						
	Dispersed CuO → Cu	Cu(II) → Cu(I)	Cu(I) → Cu	CuO (agglomeration)	Fe$_2$O$_3$ → Fe$_3$O$_4$	Fe$_3$O$_4$ → FeO	FeO → Fe
CuO	-	6.086 (280 ℃)	19.041 (309 ℃)	2.042 (385 ℃)			
Cu1Fe2	0.045 (184 ℃)	8.289 (260 ℃)	7.987 (287 ℃)	0.527 (335 ℃)	4.027 (454 ℃)	1.850 (518 ℃)	0.764 (553 ℃)
Cu1Fe1	0.023 (157 ℃)	6.265 (255 ℃)	5.518 (303 ℃)	1.284 (357 ℃)	6.025 (535 ℃)	3.991 (653 ℃)	1.677 (718 ℃)
Cu2Fe1	0.065 (149 ℃)	7.612 (236 ℃)	1.317 (261 ℃)	0.312 (332 ℃)	8.363 (473 ℃)	7.266 (580 ℃)	3.175 (647 ℃)

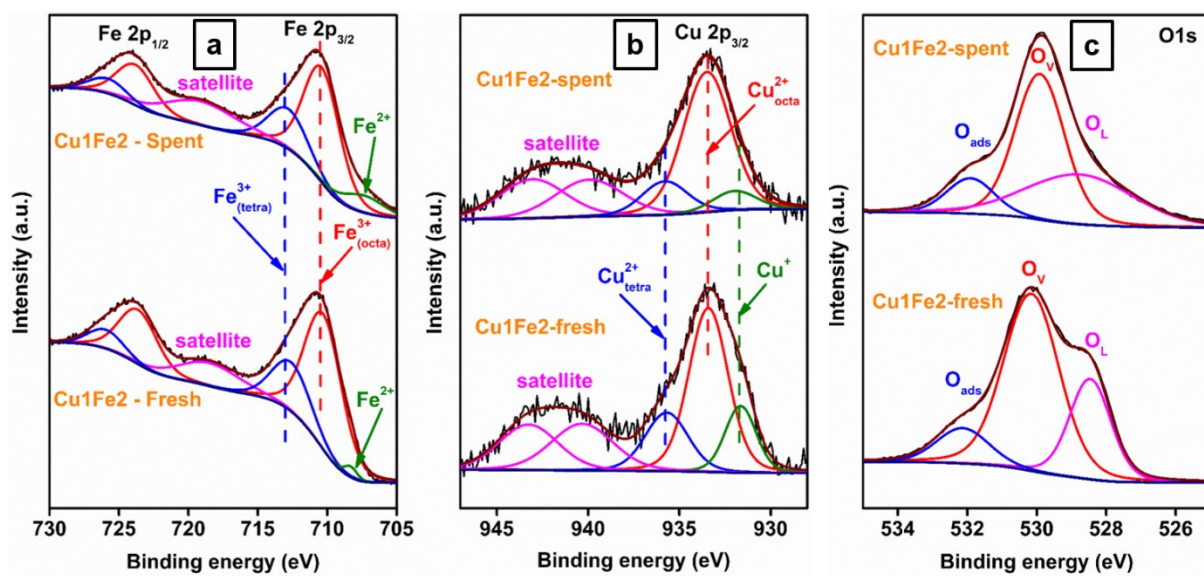* The temperature in bracket represents reduction temperature

Fig. S9 XRD pattern of spent and fresh Cu1Fe2 catalysts.
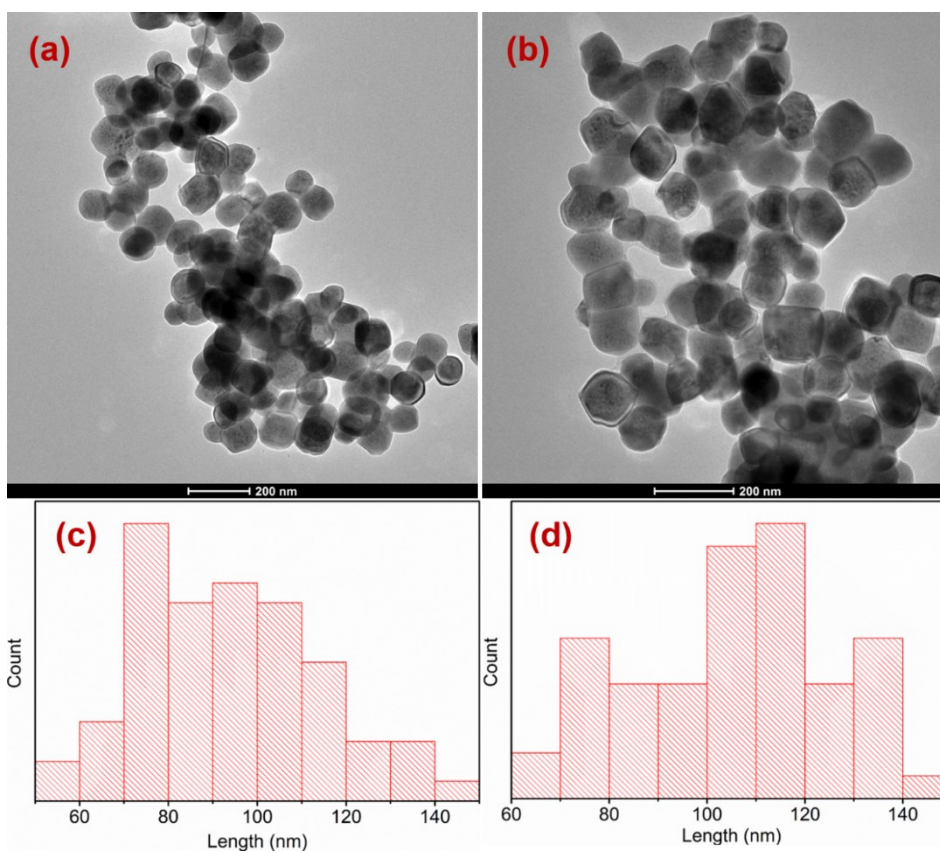
Fig. S10 TEM images of (a) fresh and (b) spent Cu1Fe2; particle size distribution of (c) fresh and (d) spent Cu1Fe2 catalysts.

Fig. S11 (a) Fe2p, (b) Cu2p, and (c) O1s XPS profiles of fresh and spent catalyst.

Calculation of ethylbenzene conversion: Analysis of the catalytic reactions was carried out by gas chromatography using an Agilent 7890B series gas chromatograph system. Conversion and product yields were estimated using anisole as an internal standard (IS).

Firstly, the response factor (RF) of the analyte was determined with respect to the internal standard (IS).

Response factors were estimated using the following equation:

$$RF = \frac{Area\ of\ IS\ \times Concentration\ of\ analyte}{Area\ of\ analyte\ \times Concentration\ of\ IS}$$

The concentration of reactant and products were calculated using the following equation:

$$Concentration\ of\ analyte = \frac{RF\ \times Concentration\ of\ IS\ \times Area\ of\ analyte}{Area\ of\ IS}$$

Conversion of the analyte was calculated using the following equation:

$$Conversion\ (\%)\ = \frac{(Initial\ concentration\ - Final\ concentration)}{Initial\ concentration} \times 100\ \%$$