Supporting Information

Deposition of Pd Nanoparticles on 2D Ni-Fe-MOF Ultrathin Nanosheets for Efficient N-Alkylation of Amines by Alcohols under Visible Light

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Figure S1. GC graphs of standard materials and the product after the reaction (a) aniline (b) benzaldehyde; (c) N-benzylideneaniline; (d) N-benzylaniline; (e) filtrate obtained after 12h of reaction over 2 wt% Pd/Ni-Fe-MOF NSs.





Figure S2. Mass spectrum of the product from aniline with benzyl alcohol.



Figure S3. Mass spectrum of the product from *p*-methoxyaniline with benzyl alcohol.



Figure S4. Mass spectrum of the product from *p*-touidine with benzyl alcohol.



Figure S5. Mass spectrum of the product from *p*-chloroaniline with benzyl alcohol.



Figure S6. Mass spectrum of the product from aniline with *p*-methoxybenzyl alcohol.



Figure S7. Mass spectrum of the product from aniline with *p*-methylbenzyl alcohol.



Figure S8. Mass spectrum of the product from aniline with *m*-methylbenzyl alcohol.



Figure S9. Mass spectrum of the product from aniline with o-methylbenzyl alcohol.

Figure S10. XPS spectra of Ni-Fe-MOF NSs (a) Fe 2p, (b) Ni 2p, and (c) S 2p.



Figure S11. FT-IR spectra of prepared Ni-Fe-MOF NSs.



Figure S12. Mott-Schottky plots of Ni-Fe-MOF NSs.



Figure S13. XRD patterns of (a) x wt% Pt/Ni-Fe-MOF NSs (x = 1, 2, 4), (b) x wt% Au /Ni-Fe-MOF NSs (x = 1, 2, 4), (c) x wt% Ag /Ni-Fe-MOF NSs (x = 1, 2, 4).



Figure S14. XRD patterns of 2 wt% Pd/bulk Ni-Fe-MOF.



Figure S15. SEM images of 2 wt% Pd/bulk Ni-Fe-MOF.



Figure S16. (a) Photoluminescence (PL) spectroscopy 2 wt% Pd/Ni-Fe-MOF NSs and 2 wt% Pd/bulk Ni-Fe-MOF, (b) Fluorescent lifetimes spectroscopy 2 wt% Pd/Ni-Fe-MOF NSs and 2 wt% Pd/bulk Ni-Fe-MOF.



Figure S17. (a) Time-dependent changes in the amounts of aniline over 2 wt% Pd/Ni-Fe-MOF NSs and 2 wt% Pd/bulk Ni-Fe-MOF in reaction systems, (b) TPD spectra of 2 wt% Pd/Ni-Fe-MOF NSs and 2 wt% Pd/bulk Ni-Fe-MOF pre-adsorbed with aniline and benzyl alcohol to saturation.



Figure S18. XRD patterns of 2 wt% Pd/Ni-Fe-MOF NSs before and after the reaction.



NH 1a	$\begin{array}{c} 2 \\ + \\ 2a \end{array} \xrightarrow{OH} \\ hv, N_2 \\ cat. \end{array}$		+		+
Entry	Catalyst	Conv.	Yield [%]		Aldehyde
		[%] ^{a)}	3 a	3b	[µmol]
1	1wt%Pt/Ni-Fe-MOF NSs	29	8	14	14
2	2wt%Pt/Ni-Fe-MOF NSs	64	26	31	59
3	4wt%Pt/Ni-Fe-MOF NSs	35	11	21	32
4	1wt%Au/Ni-Fe-MOF NSs	14	5	6	19
5	2wt% Au/Ni-Fe-MOF NSs	45	3	38	61
6	4wt% Au/Ni-Fe-MOF NSs	29	8	14	14
7	1wt% Ag/Ni-Fe-MOF NSs	17	3	8	12
8	2wt%Ag/Ni-Fe-MOF NSs	32	9	19	30
9	4wt%Ag/Ni-Fe-MOF NSs	34	9	20	30

Table S1. Light-induced catalytic performance for N-alkylation of aniline with benzyl

Reaction conditions: aniline (0.1 mmol), benzyl alcohol (2 mL), catalyst (10 mg), N_2 , 3 W bule LED visible light 12h. ^{a)} Conv. was calculated based on **1a**. alcohol under different conditions.