

Figure S1. The optical photographs of the APO and TM-doped APO-based powder samples, as indicated.



Figure S2. The XRD patterns and the least-squares fitting through the Rietveld refinement for our calcined samples as indicated.

Sample	Space group	Cell	Cell	Crystallite
		parameters	volume	size (nm)
		(Å)	(Å) ³	
APO-1000	<i>P112</i> ₁	a=5.2044	226.8	26-28
		b=5.2975		
		c=9.1935		
CoAPO-	<i>P112</i> ₁	a=5.1892	226.91	31-33
1000		b=5.2863		
		c=9.2626		
FeAPO-	<i>P112</i> ₁	a=5.1951	227.39	31-33
1000		b=5.294		
		c=9.2604		
CoFeAPO-	<i>P112</i> ₁	a=5.2043	228.82	28-30
1000		b=5.3061		
		c=9.2826		

Table S1. The structural parameters obtained from the Rietveld refinement of the XRD patterns.



Figure S3. Left panel: HRTEM of the (a) CoAPO-550, (c) FeAPO-550, and (e) CoFeAPO-550 samples pyrolyzed at 550°C. Right panel: HRTEM of the (b) CoAPO-1000, (d) FeAPO-1000, and (f) CoFeAPO-1000 samples calcined at 1000 °C.



Figure S4. The XPS spectra of the FeAPO-1000 and CoFeAPO-1000 samples. The energy edge used is also indicated.

1. <u>Calculation of Urbach energy from UV–Vis DRS</u>

Co-550: $E_u = 1/(0.8912) = 1122 \text{ meV}$ Fe-550: $E_u = 1/(1.2510) = 799 \text{ meV}$ CoFe-550: $E_u = 1/(0.8450) = 1183 \text{ meV}$ Co-1000: $E_u = 1/(16.7007) = 60 \text{ meV}$ Fe-1000: $E_u = 1/(2.4699) = 405 \text{ meV}$ CoFe-1000: $E_u = 1/(11.922) = 84 \text{ meV}$



Figure S5. Urbach energy plots for the samples calcined at 550 and 1000 °C.

Sample	%Carbon content		
	Pyrolyzed at 550 °C	Calcined at 1000 °C	
APO	5%	-	
CoAPO	7%	0.9%	
FeAPO	21%	1.3%	
CoFeAPO	13%	0.5%	

Table S2. TGA weight loss %.

2. The BET Method was used for the evaluation of the surface parameters from the adsorption-desorption isotherm.

Many unique properties are associated with mesoporous molecular sieves and measuring surface area porosity is one of the major properties for molecular sieves. The most commonly used measuring technique is the Brunauer–Emmett–Teller (BET) surface adsorption method. The surface area is major property for many applications, and it is described by the following equation:

$$\frac{1}{W((P_0/P) - 1)} = \frac{1}{W_m C} + \frac{C - 1}{W_m C} \left(\frac{P}{P_0}\right)$$

W = Weight of gas adsorbed

 $P_0/P = Relative pressure$

Wm = Weight of adsorbate as monolayer

C = BET constant

BET equation needs a linear plot of $\frac{1}{W((P_0/P)-1)}$ v/s $\frac{P}{P_0}$

Then slope (s) and the intercept (i)

$$\mathbf{S} = \frac{C-1}{W_m C} \qquad \mathbf{i} = \frac{1}{W_m C}$$

$$W_{\rm m} = \frac{1}{s+i}$$

Total surface area can be calculated from following equation:

$$\mathbf{S}_{\mathrm{t}} = \frac{W_m \, N \, A_{cs}}{M}$$

N = Avogadro's number (6.023×10^{23})

M = Molecular weight of Adsorbate

 $A_{cs} = Adsorbate cross sectional area$

Specific surface area can be determined by total surface area by sample weight according to the following:

$$S = \frac{S_t}{w}$$

In BET analysis, the pore size plays a major role because it is a very important property of mesoporous materials and is useful for many applications; moreover, it is determined from the pore volume.

The average pore radius is expressed as follows:

$$r_{\rm p} = \frac{2V_{liq}}{S}$$

Total pore volume is determined from the amount of vapor adsorbed at a relative temperature close to unity by assuming that all the pores are filled with liquid adsorbate.

 $V_{liq} = \frac{P_a V_{ads} V_m}{RT}$ $V_{ads} = Volume \text{ of gas adsorbed}$ $V_{liq} = Volume \text{ of liquid}$ $V_m = Molar \text{ volume of liquid adsorbate}$ $P_a = Ambient \text{ pressure}$

T = Ambient temperature



Figure S6. The reactor used for the photocatalytic studies. A Xe lamp was used, which is not switched on here.



Figure S7. The time-dependent conversion depicted by color change in the collected aliquots during 4-nitrophenol reduction. The numbers written on the test tubes are the time duration in minutes for which the reduction reaction was studied.



Figure S8. (a) Comparison of XRD patterns for the fresh CoAPO-1000 and recovered samples post-photocatalysis experiment. The (b) Co 2p, (c) P 2p, and (d) Al 2p XPS spectra for the recovered CoAPO-1000 sample.