Fig. S1. XRD patterns of layered precursor a) NH$_4$NiMo-a ; b) NH$_4$NiMo-b. The ◊ symbol refers to the simulated peaks from the ICSD-165342 pattern.
Fig. S2. XRD patterns for mixed oxides a) NiMo-a-ca; b) NiMo-b-ca. The α and β symbols are for α-NiMoO$_4$ (ICSD-81059) and β-NiMoO$_4$ [38] phases patterns, respectively.
Fig. S3. N₂ adsorption isotherms (left) and pore size distribution (right) for both mixed-oxides a) NiMo-a-ca; b) NiMo-b-ca
Fig. S4 N\textsubscript{2} adsorption isotherms (left) and pore size distributions (right) for both unsupported sulfides a) NiMo\textsubscript{as}; b) NiMo\textsubscript{bs}
Fig. S5. (A) The distribution for slab stacking degree of sulfides
   a) NiMo/Al$_2$O$_3$ and b) NiMo-as
Fig. S5. (B) The distribution for slab length of sulfides
a) NiMo/Al₂O₃ and b) NiMo-as
Figure S6. XANES spectra in the Ni K-edge for unsupported catalysts, ■ NiMo-as and ▲ NiMo-bs, and a Ni$_3$S$_2$ (solid line) commercial pattern.
Figure S7. Products yield vs reactant conversion for NiMo-bs bulk sulfide catalyst.

a) ☐ CHB and * BPH and first order fit (solid lines). b) ♦ cis-decalin and ▲ trans-decalin and zero order fit (solid lines).
Figure S8. Products yield vs reactant conversion for NiMo/Al₂O₃ sulfide catalyst.

a) □ CHB and * BPH and first order fit (solid lines). b) ♦ cis-decalin and ▲ trans-decalin and zero order fit (solid lines).
Figure S9. Products yield vs reactant conversion for NiMoP/Al₂O₃ sulfide catalyst.

a) □ CHB and * BPH and first order fit (solid lines). b) ♦ cis-decalin and ▲ trans-decalin and zero order fit (solid lines).